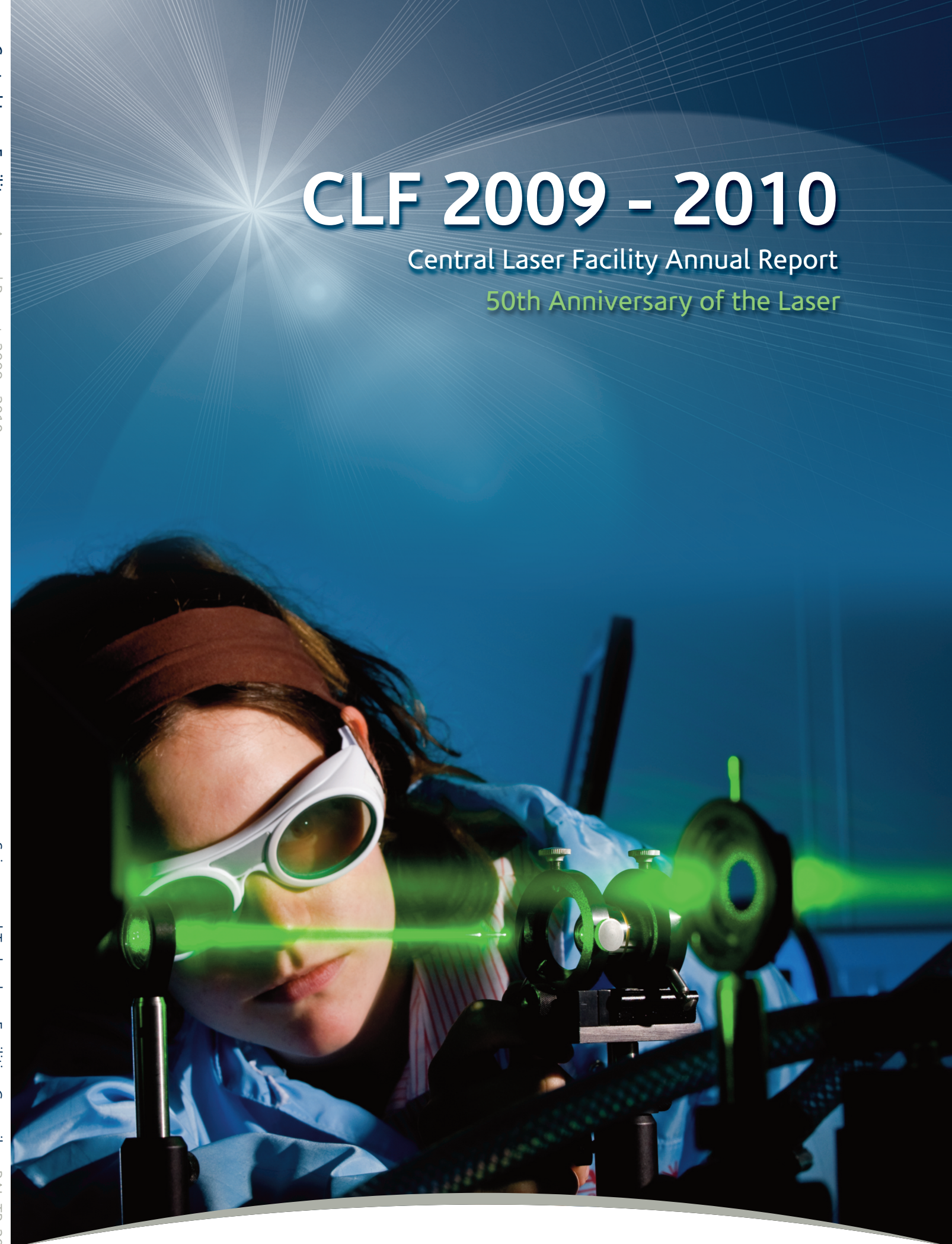


CLF 2009 - 2010

Central Laser Facility Annual Report

50th Anniversary of the Laser



CENTRAL LASER FACILITY

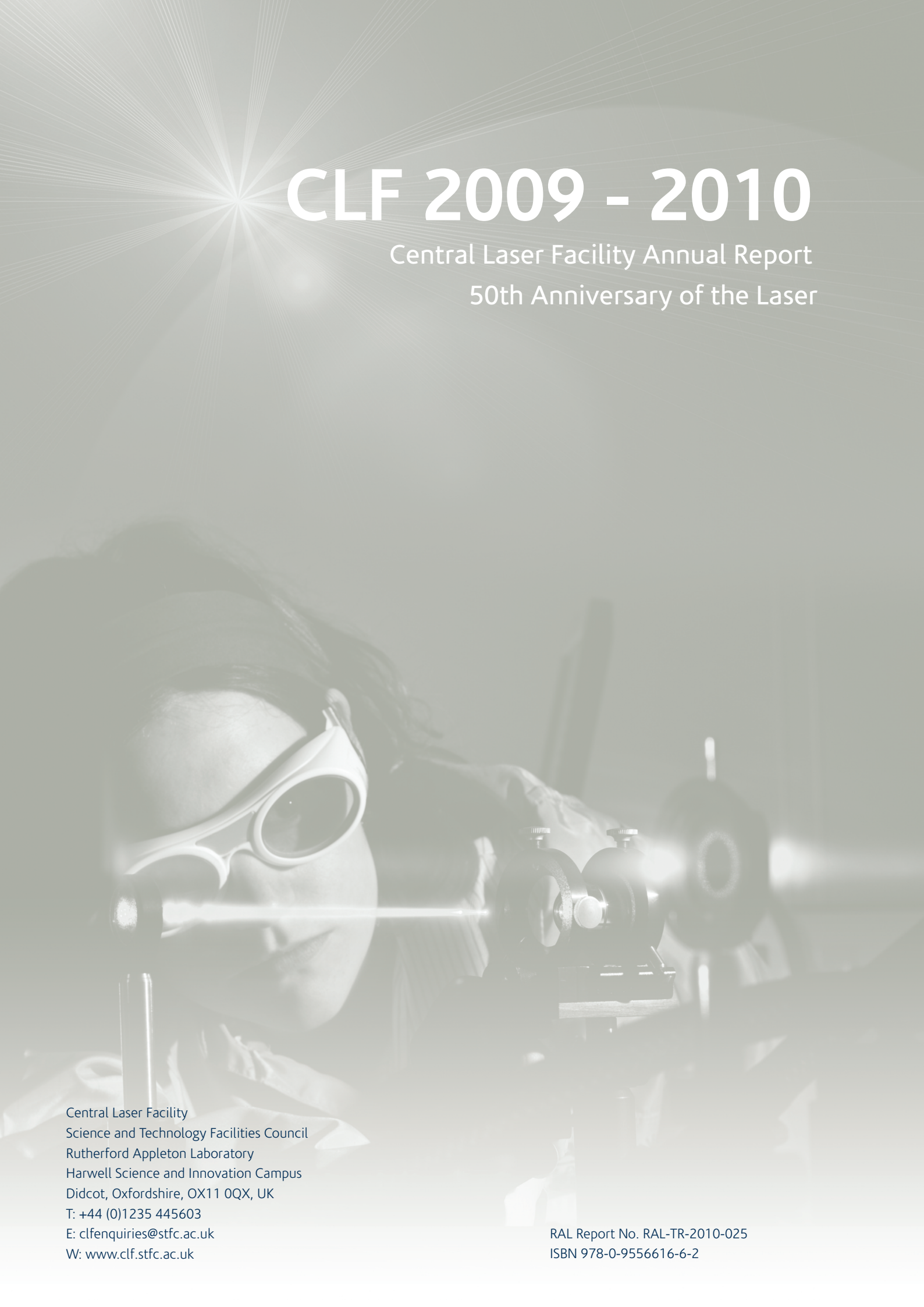
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Thanks to all the above for their contribution towards producing this report and of course to all the authors for their submissions.

Contents

Foreword	4
Overview	5

High Power Laser Science	Section 1
Plasma Physics	7
Femtosecond Pulse Physics	10
Theory and Computation	13
Ultrafast and XUV Physics	21

Lasers for Science Facility Programme	Section 2
Biology	24
Chemistry	27
Physics	31

Laser Science and Development	Section 3
Artemis	33
Astra	35
Lasers for Science Facility	38
Vulcan	41
Laser R&D	43
Target Fabrication	45
Instrumentation and Plasma Diagnostics	46

Appendices	Section 4
Schedules and Operational Statistics	47
Publications	59
Panel Membership and CLF Structure	65

CD Rom Index 	Section 5
	68

Author index	Section 6
	70

Foreword

Mike Dunne

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This annual report for the Central Laser Facility (CLF) at the STFC Rutherford Appleton Laboratory provides highlights of the scientific research which has been carried out by users of the Facility and its staff over the financial year 2009-10.

The report records an ever widening scope of research, making use of the new Artemis and LSF facilities, data from the first arm of Astra-Gemini and continuing high level output from the constantly-evolving Vulcan laser facility. This breadth of research is key to the ongoing success of the CLF – allowing continuous innovation and cross-over between areas, whilst maintaining a leading capability and the flexibility to respond to emerging new concepts and opportunities.

I am sure you will be impressed by the scale and quality of the output reflected in these pages.

Sadly, this report marks the end of my tenure as Director of the Central Laser Facility. I move on to pursue the challenge of laser fusion energy in the USA, to help contribute to the delivery of abundant clean energy – as manifested in the HiPER and LIFE projects.

Looking back at the past 5 (hugely enjoyable) years in the CLF, there were three principal aspects of our

operation which I felt needed development, building from the incredibly successful 30 years of prior research:

1. Securing a long-term future for the Lasers for Science Facility (LSF), with a clear role and fit within the research community. With the move to the new suite of laboratories in the “Research Complex at Harwell”, and a well defined new mission, I believe the LSF is now an essential component for the future scientific and economic health of physical and bio-science research at RAL.
2. Ensuring a robust upgrade programme for the CLF’s High Power Lasers. With the commissioning of Astra-Gemini and Artemis, and the decision to move forward with the construction phase of the Vulcan 10 Petawatt project, the CLF will continue for many years to provide truly world leading capability to its international user community.

3. Establishing a much stronger strategic context in which to grow and flourish. By working at the European level to build cohesion and coordination, a wholly new scale of laser research activity has been developed – manifested in the ELI and HiPER European projects. Underpinned by Laserlab-Europe and the strategic objectives of these projects, there is a robust, long-term platform to guide the CLF’s future development and demonstrate the global impact of its research.

This has all been made possible by the dedication, expertise and creativity of the CLF’s staff and our close-knit user community.

I would like to thank you all for your wonderful support, hard work and sheer brilliance over the past few years in developing and delivering ideas. The CLF can be proud to be the best in the world in its chosen fields. Long may this continue.



Professor Mike Dunne
Director, Central Laser Facility

Overview

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The CLF is a world leading centre for research using lasers. This section provides an overview of the capabilities offered to our international academic and industrial community.

Vulcan

Vulcan is a highly versatile Nd:glass laser with three independent target areas that couple long and short pulse beams in a variety of flexible geometries. A maximum of 2.5 kJ can be delivered in its eight beams. Long-pulse temporal shaping is possible, with pulse durations from 100 ps to 20 ns, and a selection of focusing, beam smoothing and harmonic conversion options. Intensities up to 10^{21} Wcm⁻² are available.

TA-Petawatt is Vulcan's highest intensity area, with a ~500 J / 500 fs beam capable of being focused to 10^{21} W/cm². It offers the capability for beam splitting, combination with a ~100 J long pulse beamline, and various probe beam options.

TA-West offers dual CPA beams coupled to the long pulse beamlines. One beam operates at 80-100 J / 1 ps (10^{20} W/cm²), and the second CPA beam operates either at 80-100 J / 1 ps, or at 500 J / 10 ps configurable in flexible geometries.

TA-East offers long pulse capability along with short pulse probe beams – in a variety of highly flexible configurations.

Work continues on a very major upgrade – to 10 Petawatt peak power (300 J / 30 fs) capable of being focused to 10^{23} W/cm² delivered to two target areas (TAP and one new one). Approval to proceed with the major build phase is expected in early 2011.

Astra Gemini

This Ti:Sapphire facility has been designed to offer 2 beams, each with a power of 0.5 PW and a repetition rate of one shot every 20 seconds. The facility will enable interaction studies up to 10^{22} Wcm⁻². F/20 and F/2 beam focusing options are available, along with a robotic solid target insertion system and a variety of 'plasma mirror' configurations for high contrast pulse delivery. The second

beam has now been commissioned, as have further improvements to the beam contrast and stability.

Artemis

Artemis offers carrier stable ultra-short pulse capability from the Infrared through to XUV spectral regions, with a tunable probe source and end-stations offering optimised time resolution or energy resolution for atomic/molecular physics and condensed matter science. The end-stations are:

- i. A high harmonic generation chamber with kHz gas-jet and XUV flat-field spectrometer (10-40 nm spectral region), together with a monochromator to select a single XUV harmonic in the spectral range 10eV-100 eV.
- ii. Materials science end-station with 2D hemispherical photo-electron analyser and <14 K five-axis cryo-manipulator.
- iii. Gas-phase end-station with a velocity-map imaging detector
- iv. A fs time and electron spin resolved end-station.

Lasers for Science Facility (LSF)

The LSF, located in the Research Complex at Harwell, clusters into two areas: molecular structural dynamics (ULTRA), and functional bio-systems imaging (OCTOPUS).

In the dynamics area ULTRA offers a state-of-the-art high power 10 kHz fs / ps system combined with OPAs to generate pulses for a range of unique pump and probe spectroscopy techniques. It provides spectral coverage from 200-12000 nm and temporal resolution down to 50 fs. This is used in the investigations of fast photodynamic processes in solids, solutions and gases. Its time resolved resonance Raman (TR³) capability enables highly fluorescent samples to be studied using a 4 ps optical Kerr shutter. The PIRATE facility (Picosecond InfraRed Absorption and Transient Excitation) gives two independently tunable beams across the mid infrared region of the spectrum for pump / probe experiments. A new development to link the two capabilities is underway (TRMPS).

In the imaging area, the Octopus cluster provides access to a broad range of capabilities from single-molecule imaging, confocal microscopy and spectroscopy, along with bio-preparation laboratories and associated tools. In addition, a 'laser tweezers' laboratory is available to study Raman spectra and pico-Newton forces between particles in solution for bioscience and environmental research.

Laser Loan Pool

Commercial laser systems are available from the EPSRC Laser Loan Pool for periods of up to 6 months at the user's home laboratory. A wide range of ancillary and diagnostic equipment is also available to support user experiments.

Engineering Services

Mechanical, electrical and computing support is provided for the operation of the laser facilities at the CLF, for the experimental programmes on these facilities and for the CLF's research and development activities. Mechanical and electrical CAD tools and workshop facilities enable a rapid response.

Theory and Modelling

The CLF will support any scheduled experiment throughout the design, analysis and interpretation phases. We offer multi-dimensional fluid, PIC, hybrid and Vlasov-Fokker-Planck modelling capabilities and access to large scale computing.

Target Preparation

A high quality target fabrication facility is operated within the CLF. This area has been significantly extended recently, offering separated characterisation, assembly, and coating areas, plus office space. It is equipped with a wide range of target production and characterisation equipment, including evaporation and sputter coating plants, interference microscopes, SEM, AFM, surface profiling and a plasma etch facility. Many targets are produced in collaboration with STFC micromachining and lithographic services. Commercial access to target preparation capabilities is available for supply to other laboratories via the spin-out company Scitech Precision Ltd.

Access to Facilities

Calls for access are made twice annually, with applications peer reviewed by external Facility Access Panels.

The CLF operates "free at the point of access", available to any UK academic or industrial group engaged in open scientific research, subject to external peer review.

European collaboration is fully open for the high power lasers, whilst European and International collaborations are also encouraged across the CLF suite for significant fractions of the time. Dedicated access to CLF facilities is awarded to European researchers via the LaserLab-Europe initiative (www.laserlab-europe.net) funded by the European Commission.

Hiring of the facilities and access to CLF expertise is also available on a commercial basis for proprietary or urgent industrial research and development.

Please visit www.clf.stfc.ac.uk for more details on all aspects of the CLF.

Plasma Physics



Plasma jets and narrow bandwidth ion spectra from thin foils irradiated at high laser intensity

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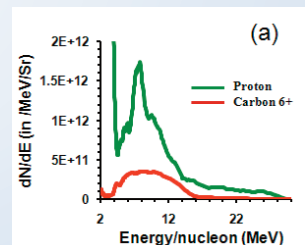
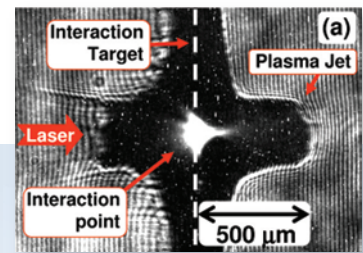
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Plasma jet evolution and ion acceleration from micron to sub-micron thickness foils following the interaction of ultra intense (up to 3×10^{20} W/cm²) laser pulses has been investigated using the Vulcan Petawatt laser. With the maximum achievable intensity on target, jets with Mach number up to 10 have been observed from few microns thick Cu foils expanding longitudinally with a velocity of 2×10^6 m/s. Using circularly polarized pulses, the uniformity of the jet density profile is improved significantly as compared to the jets observed from the interaction of a linearly polarized laser. While moderate energy (KeV) overdense plasma jets are formed efficiently for micron thickness target irradiated at 10^{20} W/cm² intensity, decreasing the target areal density or

increasing irradiance on target leads to an acceleration regime in which Carbon ions and protons are accelerated, in narrow band peaks, to tens of MeV/nucleon energies.

Experimental data showing emergence of a dense plasma jet at the rear side of the interaction target, probed transversely at 600 ps after the interaction with the high power laser (above).

Proton and carbon spectra obtained from the interaction of high power laser with 100 nm thick target.



Influence of overlapping high-intensity laser beams on electron and ion generation and transport



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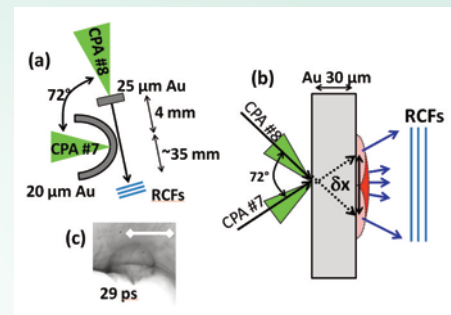
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We studied the influence of overlapping high-intensity Vulcan TAW CPA laser beams when interacting with solid metal targets. We performed an experiment using two different setups as shown in figure (a) and (b). A number of measurement techniques were used to diagnose the electron transport and proton generation, including proton radiography (see figure (c)), stacked charged particle detectors, and shadowgraphy. We clearly observed striking differences in phenomena when using, for the configurations shown in (a), two beams separated by < 3 ps, compared to having them synchronized. When they were slightly delayed, we indeed observed longitudinal parallel jet-like proton deflection patterns, likely due to transverse strong field gradients at the target rear (non-irradiated side). In the case of dual overlapping synchronized beams, as shown in (b), we observed a strongly collimated accelerated proton beam, likely to be attributed to the overlapping of bell-shaped electrostatic sheaths at the target rear surface.



Two different setups of our experiment, using either (a) two temporally separated collinear laser beams, or (b) two spatially separated, synchronised, quasi-orthogonal beams. (c) Typical proton probing data obtained in setup (a), recorded ~29 ps after the laser interaction. The bar corresponds to 500 μm in the target plane.

Characterization of a line focus back-lighter



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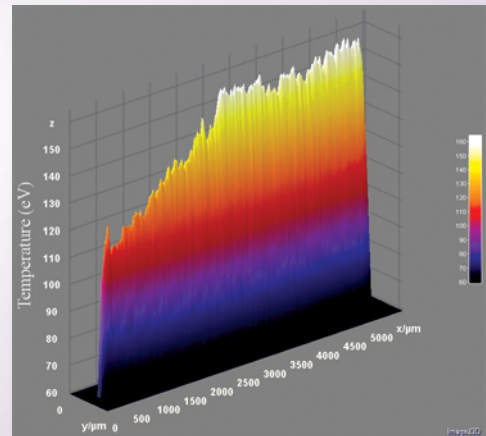
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Laser produced plasmas are often used as back-lighters to other plasmas. In order to make high quality measurements of transmission, the EUV/x-ray back-lighter source must be well characterized. Accurate information regarding the emission from the back-lighter and its transmission through the opaque plasma allows for the opacity to be effectively determined. This work demonstrates how a time and spectrally integrated image of plasma emission from a crossed-slit camera has been used to infer a spatially dependant profile of electron temperature in a line-focus EUV back-lighter.

Spatially dependant profile of electron temperature in a line-focus EUV back-lighter, inferred from a time and spectrally integrated emission profile of a crossed-slit image.



Creation of persistent, straight, 2mm long laser-driven channels in underdense plasmas for fast ignition applications



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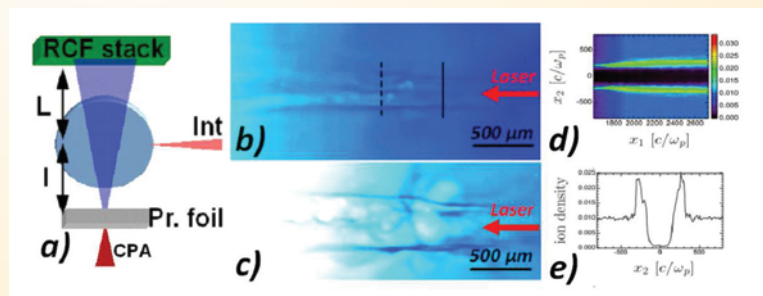
There are a number of applications of intense laser pulses that require the formation of a long-lived, long-scale, density-depleted channel in an underdense plasma. These include quality control of 100s MeV electron bunches generated in laser wakefield accelerators, high brightness X-ray generation in these channels by betatron oscillations and Fast Ignition.

Here we report the experimental detection of a smooth, persistent density depleted channel in a Deuterium plasma of initial electron density $n_e \approx 2 \times 10^{18} \text{cm}^{-3}$. The channel has been detected to extend for the entire plasma length (of the order of 2mm) and to persist, almost unperturbed, for at least 100ps after the laser has started

to propagate through the plasma. Data analysis indicate that the channel preserve sharp walls with a density accumulation of the order of 2-3 n_e and an inner density of 0.2-0.3 n_e . The experimental results have been confirmed by matching 2-dimensional Particle-In-Cell simulations.

These characteristics of the channel have been made possible by the particular set of laser parameters employed ($P_L = 6 \text{TW}$, $\tau_L = 30 \text{ps}$); the laser lays in fact below the threshold for relativistic self-focussing, thus hampering the onset of detrimental relativistic instabilities, and it is long enough to provide sustained ponderomotive push of the channel walls.

a) Top view of the experimental arrangement. b), c) Proton radiographs of the channel 100ps and 150ps after the beginning of the interaction. d) Channel creation as simulated by the 2D PIC code. e) Simulated ion density profile across the channel.



Observation of post-soliton expansion following laser propagation through an underdense plasma



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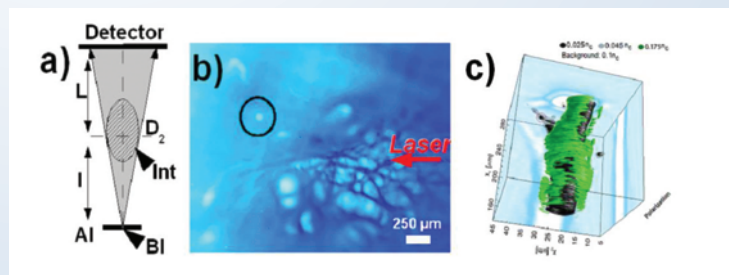
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The late-time stage of the interaction between a relativistically intense laser pulse ($E_L = 200$, $\tau_L = 30$ ps, $I_L \approx 3 \times 10^{18}$ Wcm $^{-2}$, Int laser in Figure) and a deuterium plasma with initial electron density of the order of a fraction of the critical density has been experimentally studied via the proton radiography technique. The data indicate the presence of several density depletions, located around the main laser-driven channel and at the end of the laser filaments, that are interpreted to be the late-time remnants of electromagnetic solitons (namely post-solitons). Electromagnetic solitons arise from the trapping of the laser radiation inside the plasma and, on a time scale long enough to allow ion motion, are predicted

to expand as a consequence of Coulomb explosion of the positively charged soliton core. These entities are of central relevance to this class of laser-plasma interactions, being one of the main vectors of laser energy dissipation during its propagation through the plasma.

For the first time, the temporal evolution of these structures has been experimentally measured. The data indicate deviations from a pure spherical expansion, unveiling laser polarisation effects on the post soliton dynamics. Experimental data have been found to be in good agreement with ad-hoc 3-dimensional Particle-In-Cell simulations and published analytical models.

a) Top view of the experimental arrangement. b) Typical proton radiograph of the interaction: the dark circle outlines a density bubble interpreted to be a post-soliton. c) 3D PIC simulated ion density of the interaction.



Perspectives on radiative blast waves in laser-heated clustered gases



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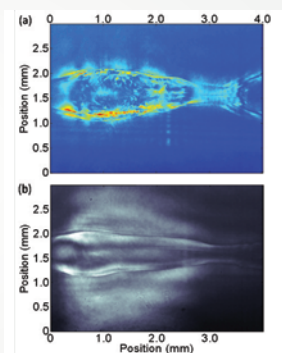
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Gases composed of clusters absorb intense laser light extremely efficiently (>90%) to form a high energy density environment (up to 10^9 Jg $^{-1}$). The subsequent explosion of the heated gas launches fast cylindrical blast waves (up to Mach 50). In high Z materials, energy losses through ionization and radiation are highly significant to the blast wave behavior. These can be categorized as optically thin radiative shocks, a regime of particular interest for laboratory astrophysics experiments. We compare the properties of radiative shocks launched in clusters to those created by other techniques which employ kJ-class laser systems. The morphology and evolution of the blast waves are sensitive to gas density because of the change in optical depth of the medium. We explore this by applying the RAPCAL code to calculate ionization levels

and radiative loss rates. We discuss the necessary conditions for radiative instabilities in the shocks and directions for future experiments.



Blast waves launched in clustered gases with a table-top laser system. These shocks exhibit turbulent structure in hydrogen (a) and large ionization precursors indicating strong radiative energy losses in xenon (b).

Femtosecond Pulse Physics

Radiation pressure effects on ion acceleration on the Gemini laser



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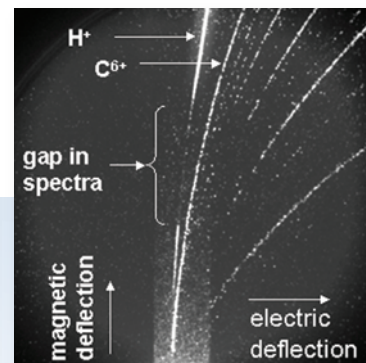
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In most laser-driven ion acceleration studies carried out to date, ions are accelerated by sheath fields established by relativistic electrons at target surfaces, via the so-called Target Normal Sheath Acceleration (TNSA). A separate mechanism, Radiation Pressure Acceleration (RPA), has attracted extensive theoretical attention in recent years. Radiation pressure is exerted at the laser reflection point on a foil surface, resulting in local electron-ion displacement, and ion acceleration via the ensuing space-charge field. Cyclical reacceleration of the target ions in the Light Sail RPA mode accessible with ultrathin foils is predicted to lead to high acceleration efficiencies, and produce



Spectrum of C^{6+} and H^+ ions, showing a proton peak at the high energy end.

energetic, narrow band ion beams. In an experiment carried out on the Astra-Gemini laser we detected features consistent with the onset of this new acceleration mechanism. Spectral peaks as shown in figure were observed for very thin foils and for circularly polarized pulses, and simulations indicate their origin from RPA.

Ion acceleration from foil targets in the ultraintense ultrahigh contrast regime



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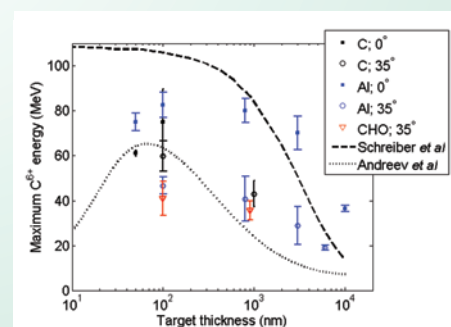
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An experiment was conducted on the Astra-Gemini laser with one of the main objectives being to investigate the acceleration of carbon ions at the highest laser intensity achievable today. The Astra-Gemini laser delivers pulses of duration ~50fs with energies up to 12J at a wavelength of 800nm, reaching intensities $\sim 10^{21} \text{ Wcm}^{-2}$. We explore the TNSA acceleration mechanism using a range of target materials and thicknesses as well as investigating the effect of the laser

incidence angle. We found that the TNSA mechanism gives maximum ion energy for a target thickness around 100nm independent of its composition. The highest carbon ion energies were produced from pure carbon targets. Our most interesting result is the first observation of an increase in ion energy when the incidence angle of the laser is changed from 0° to 35° , in contrast to previous work.



Average of the maximum C^{6+} ion energy over multiple shots as a function of target thickness for different materials. The errors bars represent the standard deviation of the maximum energy.

Characterisation of debris emission from thick targets on Astra-Gemini



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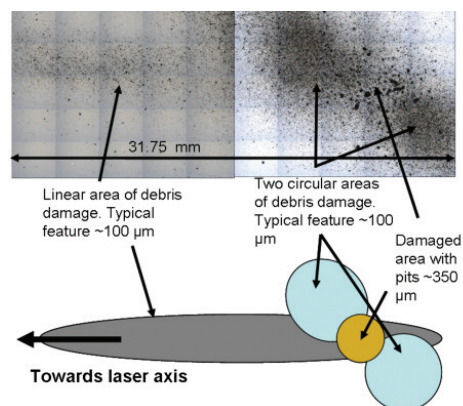
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Debris characterisation measurements were made during radiation commissioning of Astra-Gemini. During the commissioning the distribution of debris from thick targets was studied. The targets used were 0.5 mm Cu, CH and Ta and 50 μm Au. The diagnostics used to measure the debris distribution were glass witness plates positioned around the target. For the 0.5 mm thick targets it was found that the debris was mainly distributed along the rear surface target normal and laser directions with particulates typically of around 400 μm . For the 50 μm thick target the distribution was mainly along the front and rear target normal directions with particulate sizes of less than 100 μm . Damage to the witness plate surface was also observed for 50 μm thick targets along



Debris damage to glass witness plate at the target normal rear surface position. The images at the top are scans of the damage to the plate surface and the bottom diagram is a schematic of features found in the damage distribution.

the rear target normal direction. This indicates that target thickness has a significant effect on debris production and distribution.

High rep rate Thomson parabola-MCP assembly for multi-MeV ion spectroscopy



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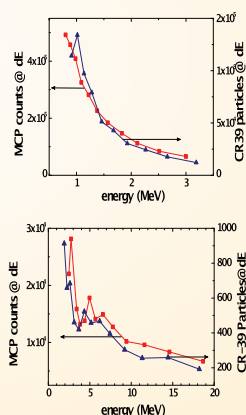
C.M. Brenner, P.S. Foster, P.L. Gallegos,
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The absolute calibration of a micro channel plate (MCP) detector, installed as detector in a Thomson parabola spectrometer has been carried out. The calibration delivers the relation between a registered count numbers in the CCD camera (on which the MCP phosphor screen is imaged) as a result of the impact of an ion beam onto the MCP. The particle response of the whole detection system was evaluated by using laser accelerated ions with proton energies up to 3 MeV and carbon ion energies up to 16 MeV. In order to obtain an absolute measurement of the number of ions incident on the MCP detector, slotted CR-39 track detector was installed in front of the MCP. The signal registered on the MCP due to ions propagating through the CR-39 slots is compared to the number of particles counted on the adjacent CR-39 stripes after the etching. Careful consideration is required in evaluating the response of the MCP, since in the

arrangement employed different energy ions are incident on the MCP at different angles (unlike, for example, ions accelerated by linear accelerators), and this may affect the gain and secondary electron yield. For this reason, an in-situ calibration is important for a correct data analysis.



Correlation of integrated counts in dE energy interval due to MCP signal with number of particles on CR-39 in dE energy interval plotted with respect to energy for Protons and C^{6+} .

Proton/ion energy scaling and laser conversion efficiency using 50fs, 10^{20} - 10^{21} W/cm² Astra-Gemini pulses



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K.E. Quinn, L. Romagnani, M. Zepf,
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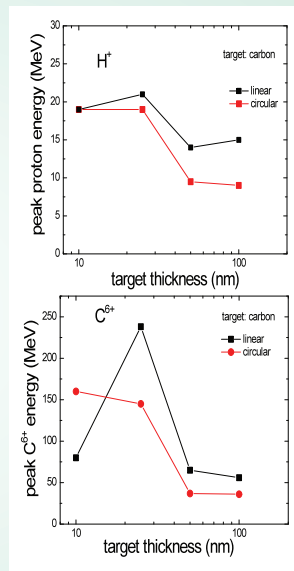
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Experimental investigations of ion acceleration from thin foil targets irradiated with ultra-short (~ 50 fs), high contrast ($\sim 10^{10}$) and ultra-intense (up to 10^{21} W/cm²) laser pulses on Astra-Gemini laser has been performed. These measurements provided for the first time the opportunity to extend the scaling laws for the acceleration process in the ultra-short (tens of fs) regime beyond the 10^{20} W/cm² threshold. The scaling of accelerated ion energies was investigated by varying a number of parameters such as target thickness (down to 10 nm), laser light polarization (circular and linear), angle of laser incidence (oblique- 35° , normal) and laser energy. Maximum proton energy >20 MeV and C^{6+} energy ~ 235 MeV has been observed. The effect of target thickness on the ion flux produced was also investigated at 35° and normal laser incidence on target.



Maximum proton/ion energy dependence on target thickness at normal incidence on aluminium target for linear and circular polarisations

Investigation of contrast of Astra-Gemini

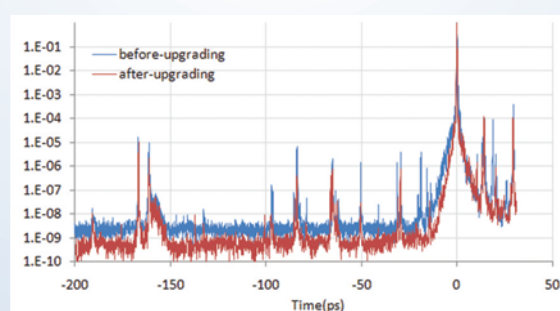


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The temporal contrast of the laser pulse of extreme power up to 10^{22} W/cm² plays a crucial role in the high-field laser-matter interaction experiments as the excessive prepulse intensity can significantly affect experiment conditions due to preplasma formation. To meet an increasing demand for higher temporal quality of laser pulse, the temporal profile of Astra-Gemini was extensively investigated under various conditions, using a 3rd-order auto-correlator. The effect of various techniques has been investigated, including the spatial

filter, aperture and minimising the scattering and loss etc. To minimise the overall ASE noise level, the front-end was upgraded and optimized to suppress the intracavity ASE noise, providing a cleaner seed pulse by a factor of ~ 10 . As a result, the overall incoherent ASE and coherent contrast was improved by one order of magnitude as seen below. Some of the replica pre-pulses was reduced by a factor of ~ 100 or eliminated by using components with higher quality antireflection coatings.



Overall contrast before and after front-end upgrading.

Theory and Computation

Electron transport during shock ignition



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In the 'shock ignition' approach to Inertial Fusion Energy (IFE), the fusion target is compressed at a relatively low temperature and then ignited by a high-pressure shock driven into the target by a short high-intensity laser pulse. Because of steep temperature gradients, electrons which transport energy into the target to drive the shock must be modeled kinetically by the Vlasov-Fokker-Planck equation. The laser-target interaction also does not have

time to relax hydrodynamically to the steady ablation profiles characteristic of the compression phase. Instead, shock ignition lies at the boundary between the 'ablation' regime and a 'supersonic heat front' regime in which high energy electrons stream freely into the target. High energy electrons transport energy deep into the compressed target where they beneficially generate high pressure at high density.

New perspectives on the electrodynamics of intense laser-plasmas



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A summary is given of work undertaken over the past twelve months by the Lancaster Mathematical Physics Group and Cockcroft Institute that focusses on new avenues for theoretical exploration of the behaviour of intense laser-plasmas. Items covered include an examination of Born-Infeld electrodynamics using plasmas,

new geometrical analyses of relativistic collisional and collisionless plasmas including a new method for analysing Landau damping in non-stationary and inhomogeneous relativistic plasmas, and a new investigation of the interaction of electromagnetic fields with accelerating matter.

Cavitation and shock wave formation in dense plasmas by relativistic electron beams



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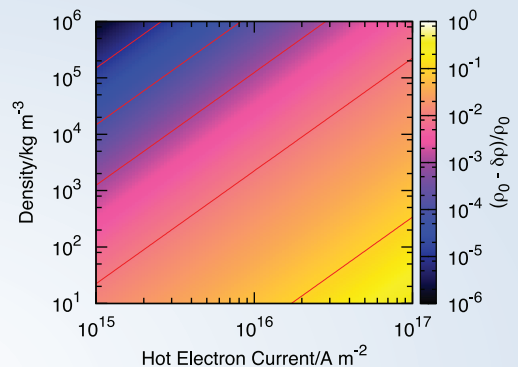
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The propagation of a high current relativistic electron beam through dense plasmas, for example in fast-ignition inertial confinement fusion, produces strong heating and magnetic field generation. The $\mathbf{j} \times \mathbf{B}$ force and thermal pressure gradient that the return current creates may in fact cavitate and cause shock waves in the plasma around the electron beam.

Work has been undertaken to investigate this effect in different regimes of plasma density and hot electron current. An analytic model has been developed that gives good estimates of the density, pressure, magnetic field and velocity obtained in the plasma. This model is compared against the results from an MHD code that includes the effects of resistive field growth, Ohmic heating and the $\mathbf{j} \times \mathbf{B}$ force. The strength of the cavitation is



A parameter scan across a range of hot electron current density and initial plasma mass density. The colour scale shows the fractional amount of cavitation expected from the analytic model.

found to be dependent upon the ratio between j^2 and the initial mass density. It was found that cavitation is indeed relevant to fast-ignition, and is strong enough to launch shocks in certain circumstances.

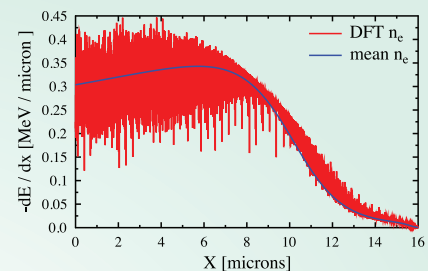
Alpha-particle stopping power for dense hydrogen plasmas



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For beam energies from 0–20 MeV, a good agreement is obtained between calculated ion ranges for α -particles stopping in dense hydrogen for both highly fluctuating electron densities from DFT-MD and uniform averaged density profiles. In addition, the range calculations demonstrate good agreement with analytical range calculations using stopping power approximations in the limit of both the high and low kinetic energy as well. This agreement implies that for calculations of large-scale effects, such as ion range or averaged energy deposition, the increased accuracy provided by applying a DFT-MD density profile has little effect on the final result. It is also observed that the general functional form of the Bragg curves calculated for the variable



Comparison of α -particle stopping in hydrogen using a density profile obtained by DFT-MD and a constant mean density. The beam energy is $E = 3.5\text{MeV}$. The plasma has a temperature of $T=20000\text{K}$ and a mean electron density of $n_e = 1.661 \times 10^{24} \text{ cm}^{-3}$.

and uniform density cases is similar. However, in variable density DFT-MD cases, fluctuations in density cause dramatic changes in the calculated stopping power over small spatial scales. This huge straggling must be considered in situations with small fluxes where heating cannot be averaged over many particles.

An object-oriented 3D view-factor code for hohlraum modelling



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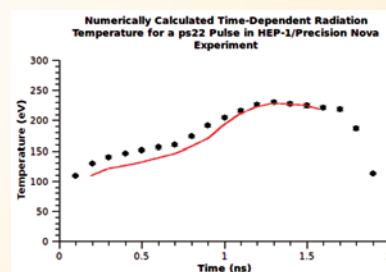
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An easy to use 3D view-factor code has been developed for use in high power laser experiment design. View-factor codes use simple arguments to determine the quantities of radiation that can flow through a hohlraum geometry, and then solve a power balance equation to calculate the spatial distribution of radiation throughout a target. Because of the relative speed involved in setting up and running simulations, 3D view-factor codes are especially suited to prototyping experimental target designs. The code allows the user to build up a wide range of hohlraum designs by combining a number of geometric primitives. Drive laser configurations can be specified in terms of individual beams or in rings; the beams themselves are implemented via a self-adjusting ray-tracing algorithm. The code has been tested against results previously published and obtained by other experimental groups, and has been shown to reproduce experimental results with reasonable accuracy.



A cut-away rendering of a Nova hohlraum geometry created by the code.



Time-dependent temperature measurements from HEP-1/Precision Nova shots (continuous line) compare well with numerical calculations produced by the code.

Calculation of Siegert states in electric field: from atoms to molecules



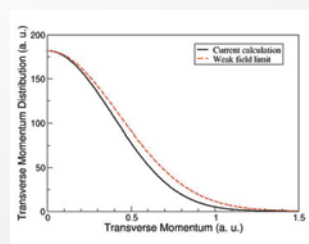
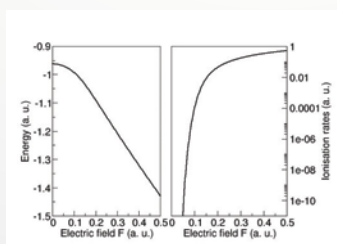
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S. Watanabe (The University
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We propose a numerical method to calculate the Siegert states of molecules in a static electric field which are solutions of the stationary Schrödinger equation satisfying the regularity and outgoing-wave Siegert boundary conditions. This work follows the one for atomic Siegert states in the reference, where they calculate complex eigenenergies and eigenfunctions for one-electron atomic potential. Here we describe the modifications necessary to obtain such quantities in complex

molecular cases. We discuss the reduced number of symmetries in the molecular problem and the inclusion of m -coupling. The two figures below present the result obtained for the lowest sigma state of H_2^+ with the soft Coulomb approximation for the electric field of 0.5 a.u.. The molecule axis is parallel to the laser field.

P. A. Batishchev, O.I. Tolstikhin and T. Morishita, submitted.



Finite size effects in high-intensity QED



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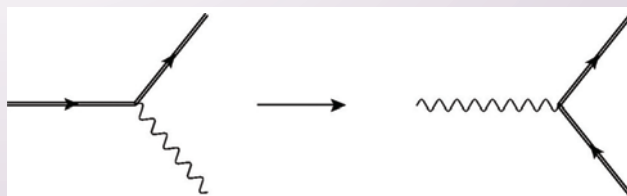
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With laser fields approaching magnitudes close to the Sauter-Schwinger limit the use of strong-field quantum electrodynamics becomes mandatory. The latter is based on electrons 'dressed' by plane wave fields, solely dependent on an invariant phase. The theory developed in the sixties assumed infinite spatio-temporal extent which at that time seemed a good approximation as the number of cycles per pulse was large compared to unity. Nowadays, however, at ultra-high intensities, pulses are ultra-short, and

effects of finite pulse duration must be taken into account.

We find that for both nonlinear Compton scattering and stimulated pair production finite size effects tend to 'wash out' the clear spectral signals caused by long-time averaging which is no longer possible for short pulses. Hence, observation of intensity effects on emission and production spectra clearly will require experimental fine-tuning.

Nonlinear Compton scattering or photon emission (left) and stimulated pair production (right). The processes are related by crossing symmetry. Both the photon emission spectrum and pair production rate are rather sensitive to the details of the laser beam, in particular its pulse duration.



Multi-electron dynamics on the femtosecond time-scale

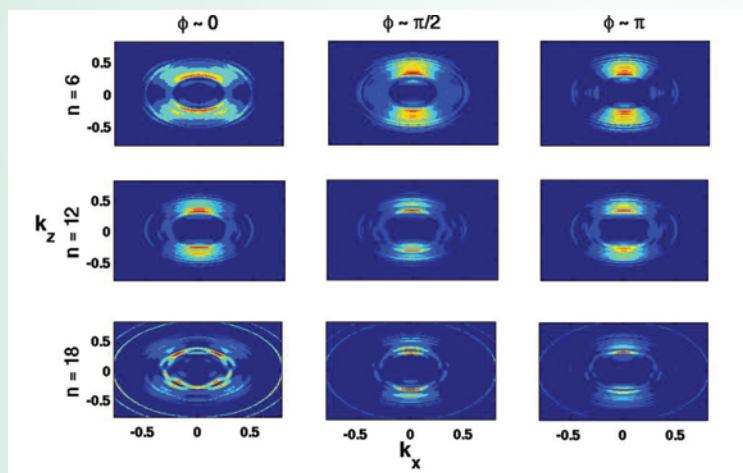


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The recent emergence of attosecond light sources promises to provide deeper insight into ultrafast correlated dynamics between electrons in complex matter. Using time-dependent R-matrix theory we have been able to investigate such dynamics within an atomic system.

We consider C^+ in its ground state with magnetic quantum number $M=0$. The ion is excited by a linearly polarized XUV pulse into a superposition of the $2s2p^2\ ^2S$ and 2D states. The repulsion between the two $2p$ electrons results in interference between the two states which occurs on a time-scale of 1-2 femtoseconds. By subsequently ionizing C^+ with a delayed ultrashort pulse and analyzing the properties of the ejected electron we are able to probe the interference and investigate the role of electron-electron interactions in the transition from ultrashort pulse excitation to long-pulse excitation.



2-dimensional momentum distributions of an electron ejected from C^+ obtained for three different excitation pulse durations: $n = 6, 12$ and 18 cycles. The plot shows a breathing motion for both $2p$ electrons for excitation pulse lengths of 6 and 18 cycles. ϕ corresponds to the phase of the breathing motion.

Hydrodynamics of the Yukawa one component plasma



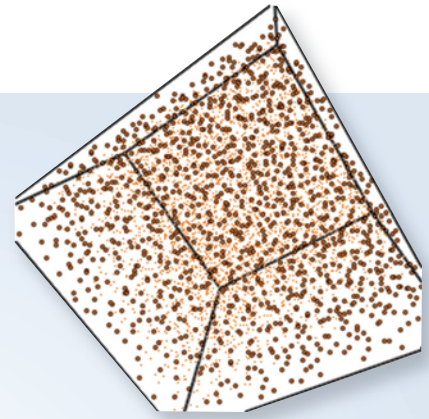
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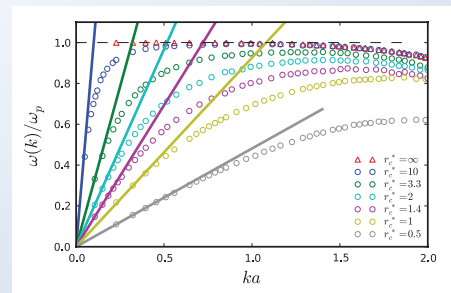
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The hydrodynamic description is a very common starting point for investigating a wide range of basic and complex physical phenomena in fluids. We investigate using Molecular Dynamics simulations when this macroscopic description (rather than a more detailed microscopic description) is applicable for a physical system. To achieve this we consider the microscopic dynamics of the so-called Yukawa One Component Plasma model. In particular, we determine how the domain of validity of the hydrodynamic description varies as the level of many body correlations (i.e. non-ideality) is varied.

Brillouin peak dispersion relation for interaction potentials of various ranges against the hydrodynamic (linear) predictions.



Our results elucidate the nature of the hydrodynamic limit when many body correlations are present, particularly for systems with long ranged interactions. They can be used to model ion dynamics in dense plasmas (e.g. 'Warm Dense Matter').



The accuracy of rear-surface measurements of the angular-divergence of a laser-generated fast-electron beam

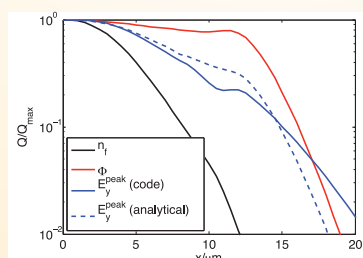


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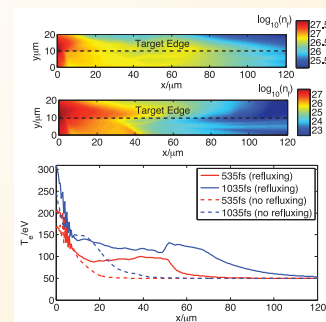
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The interaction between high-intensity lasers and solid targets is of key importance in fast-ignition inertial confinement fusion, which may allow fusion ignition with less laser driver energy; and ion-acceleration, which has potential applications to cancer therapy. Understanding the properties of the relativistic electron beam generated in these interactions is crucial. In particular, its angular divergence has ramifications for the aforementioned practical applications. This is often diagnosed using the temperature profile or the size of the region over which ion are emitted on the target's rear-surface.

Simulations performed with the hybrid Vlasov code FIDO demonstrate that re-circulating currents inside the target lead to a broadening of the temperature hot-spot on the rear, thus over-estimating the inferred angular divergence of the fast-electron beam. The sheath potential was observed to be weakly dependent on the beam density and so the ion-spot size is a poor measure of the angular divergence.



Fast-electron number density, peak sheath field and sheath potential along the target's rear-surface after 100fs, and E_y^{peak} (analytical) $\propto n_i^{1/2}$.



Fast-electron number density with refluxing (top) and without refluxing (middle) after 1035fs. Background temperature at the rear-surface (bottom).

High absorption of ultra-intense laser pulses in near critical plasmas

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Of all the problems that ultra-intense laser-plasma physics is concerned with, the coupling of energy from the laser pulse into the kinetic energy of the plasma particles is perhaps the most important. Attaining efficient coupling is highly important and even critical to a number of prospective applications. Fast Ignition ICF and laser-driven ion acceleration are examples of such applications where coupling efficiency is either a very important or critical issue.

In the work reported here, we analyzed the absorption of 30-100fs, $\lambda \sim 1\mu\text{m}$, $a_0 \gg 1$ laser pulses in near-critical plasmas. By this we mean plasmas are relativistically transparent and span the density range $0.1n_c < n_e < a_0 n_c$, where n_c is the non-relativistic critical density.

From energy considerations we have derived new analytic formulae for the two mechanisms of absorption in near-critical plasmas: Leading Edge Depletion (LED) and Transverse Ponderomotive Acceleration



(TPA). For LED, for example, we obtained the following new formulae for the leading-edge propagation velocity:

$$u = \frac{v_g}{\frac{n_e}{2n_c} + 1}$$

Our new formulae have been rigorously compared to a large number of 1D and 2D fully electromagnetic Particle-in-Cell simulations.

This study has thus shown that the two absorption mechanisms that have been suggested for interactions with near-critical plasmas can both occur separately in different regions of parameter space, and, where they do, their gross properties are well described by the new analytic formulae derived here. Since LED is a significantly faster absorption mechanism than TPA this is an important step towards optimizing absorption in the near-critical regime.

Modelling Spitzer transport in laser produced plasmas by direct Vlasov methods with a BGK collision operator



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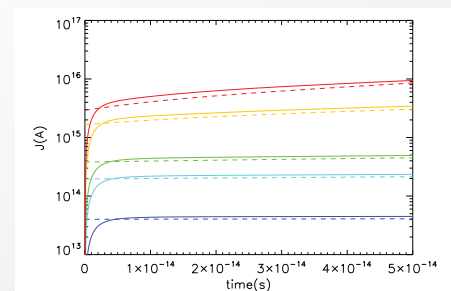
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In short-pulse laser matter interaction, the absorption of incident laser energy is dominated by collisionless mechanisms but the onward transport of these fast moving electrons, and the resultant heating of the target, is dependent on the response of the cold, resistive, background plasma.

Here we briefly outline a simple approach to including collisional physics in the direct Vlasov solver VALIS, based on the BGK

(Bhatnagar, Gross and Krook) collision operator. We are able to demonstrate the efficacy of this approach in two key transport problems in laser plasma interaction: thermal conduction and electrical resistivity.

Comparison of Spitzer currents (dashed lines) with those calculated using VALIS with a BGK collision operator (solid lines) for external fields of: 10^7Vm^{-1} (blue); $5 \times 10^7\text{Vm}^{-1}$ (cyan); 10^8Vm^{-1} (green); $5 \times 10^8\text{Vm}^{-1}$ (yellow); and 10^9Vm^{-1} (red), for the case of both electron-electron and electron-ion collisions.



Effect of channel profile evolution on laser-driven electron acceleration in plasma channels

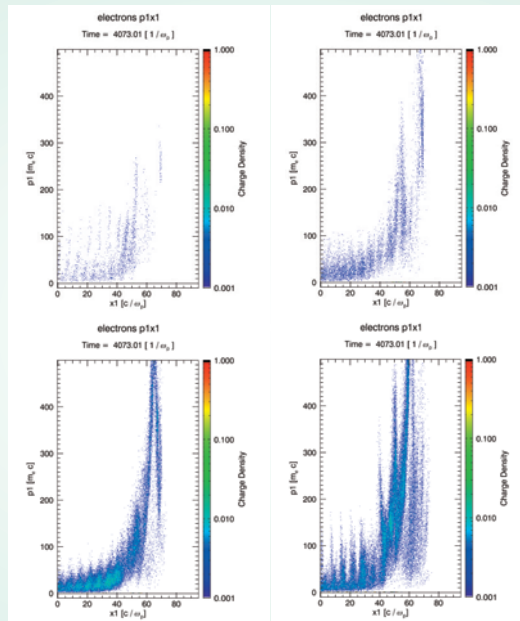


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The trapping and acceleration of electrons by a laser pulse propagating through a plasma channel has been investigated numerically. The influence of the laser intensity, background plasma density, channel transverse profile and degree of ionisation of the plasma was studied. It was found that the laser intensity and background density have the most significant impact on electron trapping, while the degree of ionisation has hardly any influence at all.

Electron trapping in a laser-driven wakefield, versus laser intensity. Laser amplitude a_0 is 1.19 (top left), 1.68 (top right), 2.0 (bottom left), 2.5 (bottom right), all other parameters equal. Laser spot diameter is 10 micron, wave length is 800 nm, $\omega_0/\omega_p = 13.2$, pulse duration is 50 fs. An increased laser intensity leads to enhanced electron trapping and acceleration.

A Vlasov-Fokker-Planck code for shock ignition

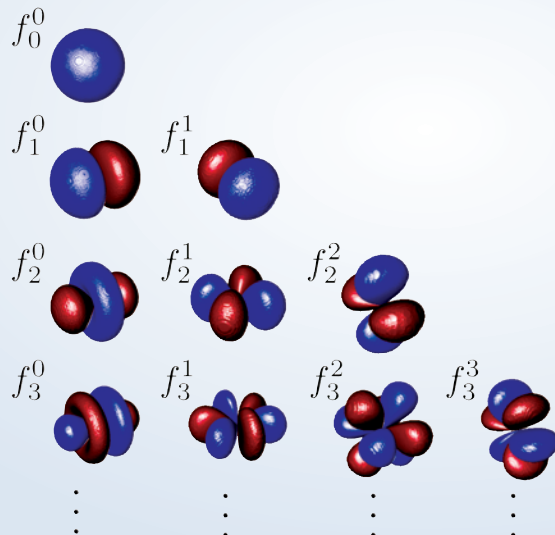


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A 2D3P parallel object-oriented Vlasov-Fokker-Planck code that relies on the expansion of the electron distribution function to spherical harmonics has been developed, in order to study non-local electron transport for Shock Ignition.

The code makes use of a rigorous formalism for the collisions between electrons, which derives from the Rosenbluth potentials and conserves energy and number. This code makes it possible to accurately model the kinetic as well as the hydrodynamic behaviour of the plasma and is particularly efficient for collisional plasmas. For Shock Ignition the electron temperatures range from more than 100keV to 10eV while densities range from less than critical to greater than solid. Shock Ignition is therefore an excellent candidate for this VFP code, because the target is sufficiently collisional to allow for extremely efficient modelling.



The first ten spherical harmonics.

Multi-component effects on the x-ray scattering signal from warm dense matter



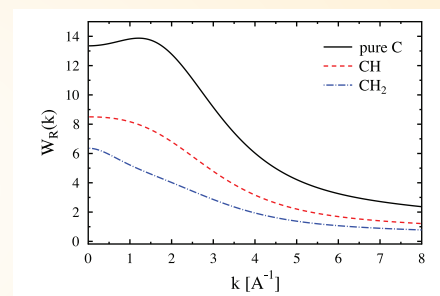
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In this contribution, we investigate the effects of multiple ion species on the x-ray scattering process from warm dense matter. In particular, we discuss elastic scattering, that is, the weight of the Rayleigh peak. Based on partial structure factors from hypernetted chain solutions, a generalised approach of the theoretical model is applied to account for multi-component effects. We demonstrate that mutual correlations significantly influence the partial structure factors due to the fact that the ions with the highest charge imprint their structure on the other components. The weight of the Rayleigh peak that is directly related to the static structure factors is thus also sensitive to the interplay between the different correlated ions in the systems. These effects are especially pronounced in the case of forward scattering, i.e. for small k values. Furthermore, we demonstrate that the full multi-component description is also necessary for cases where x-ray

scattering is dominated by one species. This effect is related to the differences in the statistical weight of the different contributions to the Rayleigh peak.



Comparison of the weight of the Rayleigh peak for warm dense carbon, CH and CH₂. The carbon density of $n_C = 5 \cdot 10^{22} \text{ cm}^{-3}$, the temperature of $T = 8 \text{ eV}$ and a charge state of $Z_C = 2$ are fixed for all systems. For the calculation of CH and CH₂, fully ionised hydrogen, i.e. $Z_H = 1$, with densities of $n_H = 5 \cdot 10^{22} \text{ cm}^{-3}$ and $n_H = 10^{23} \text{ cm}^{-3}$ is applied, respectively.

Ultrafast and XUV Science

Quasi-classical model of non-destructive wavepacket manipulation by intense few-cycle nonresonant laser pulses

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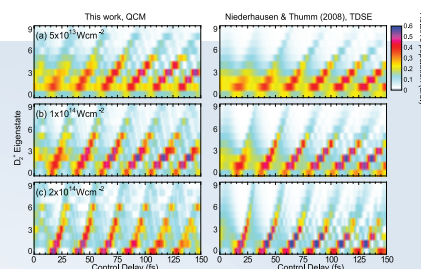
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Matter exposed to strong-field few-cycle laser pulses is violently distorted on a timescale comparable to vibrational and electronic motions. In molecules, the resulting electric field polarizes electron orbitals to the extent that ionization and Coulomb explosion are likely, without the requirement of photon energy resonance. By pumping D_2 with an intense few-cycle (~ 10 fs) pulse, ionization launches a vibrational wavepacket in D_2^+ which may be imaged some time later by a similar probe pulse; such wavepackets have been experimentally observed.

A few-cycle "control" pulse arriving between the pump and probe applies a dipole force while the wavepacket is propagating, transferring energy into or out of the system, causing a redistribution of vibrational population. This quasi-classical



Final vibrational state population distributions for three different control pulse intensities as the temporal separation between the pump and control pulse is varied. The results of Niederhausen and Thumm, Phys. Rev. A 77 013407 (2008) are reproduced for comparison with the QCM, which reproduces the periodicity and relative shift of vibrational population as the delay between the pump (6 fs, 10^{14} Wcm $^{-2}$) and the control is varied.

model (QCM) describes the modification of the wavepacket by simulating an ensemble of classical trajectories evolving on the dynamically-distorted potential energy of the system. Manipulating the internal states of molecules has implications for quantum computation, attosecond electronic state observation and the formation of a molecular quantum gas.

Monochromatic XUV-photon + strong-field NIR cross-correlation by atomic excitation and ionization

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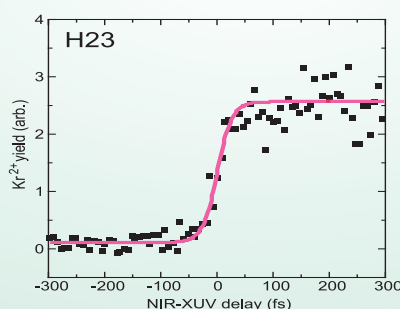
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State-of-the-art x-ray free electron lasers and attosecond high harmonic generation (HHG) sources are being used to probe electronic dynamics under on ultrashort time-scales and high intensities. In a novel cross-correlation experiment, we demonstrate the capabilities of the recently commissioned Artemis facility which neatly combines the advantageous characteristics of XFELs and attosecond

sources – energy tunability, sub-cycle synchronization to an optical laser and ultrashort pulse duration.

An intense 30 femtosecond near-infrared (NIR) laser pulse is split 3:1. The low energy pulse produces a broad spectrum of XUV photons through HHG in a gas jet, which are monochromated allowing a single 35.9 eV harmonic to be focused into a krypton target, generating highly excited Kr^+ ions through single-photon absorption. The high energy NIR pulse is also focused into the krypton to a high intensity, and the delay between NIR and XUV can be controlled with sub-cycle resolution. Depending on the arrival times of the XUV and NIR, enhancement of the Kr_2^+ ion yield is used to measure the XUV pulse duration, performing an atomic cross-correlation.



Measured and predicted Kr_2^+ yield as a function of NIR-XUV delay for harmonic 23 (photon energy 35.9 eV, wavelength 34.6 nm). The best least-squares fit reveals the rise of the yield is the result of H23 having a duration of 24 ± 2 fs.

Multi-pulse scheme for controlling electron localisation upon molecular dissociation



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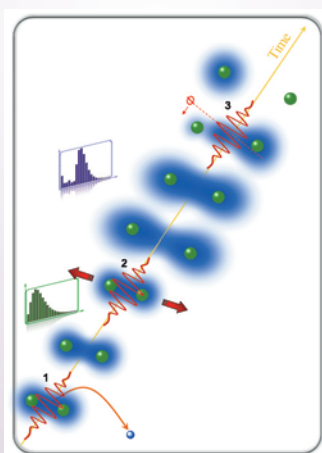
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Quantum control of chemical reactions in molecules requires the precise manipulation of electronic wavepackets, which typically evolve on attosecond timescales. A powerful technique for controlling the electronic dynamics during molecular dissociation is to use few-cycle femtosecond laser pulses of infra-red radiation.

Through controlling the relative phase relationship, ϕ , between the carrier field and pulse envelope, and using a sequence of such pulses, a new scheme has been identified for directing the localisation of the electron upon molecular break-up, i.e. whether the electron goes to the 'left' or 'right' nucleus.

This ultrafast scheme has been identified using quantum simulations of the deuterium molecular ion (D_2^+) exposed to a sequence of 7 fs pulses. With carefully chosen pulse parameters it is possible to optimally control the vibrational wavepacket dynamics and the final dissociation event, resulting in a final 'left:right' ratio of the electron localization which exceeds 10:1.

Proposed multi-pulse scheme: Pulse 1 ionises a D_2 target launching a coherent D_2^+ vibrational wavepacket. Pulse 2 non-destructively manipulates the bound wavepacket into a new vibrational distribution. Pulse 3 then dissociates the molecular ion, where careful choice of delay time allows the dissociation to be maximised and tuning of ϕ leads to strong localization of the electron to a particular nucleus.



Intensity-resolved ionization processes in few-cycle strong-field laser pulses



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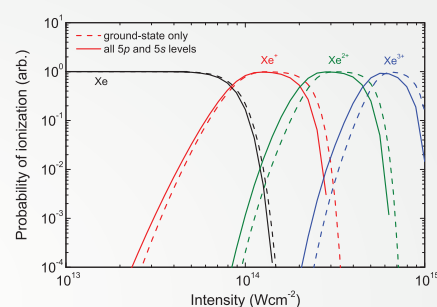
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Few-cycle strong-field laser pulses cause tunnel ionization of atoms and molecules by violently distorting the electron orbitals. It is commonly assumed that tunnelling leaves the ion in the ground-state. However, enhanced ionization yields indicate excitation processes which populate lower-lying energy levels. Alternatively, these levels could be populated directly by tunnelling. Removal of electrons from lower-lying orbitals by tunnelling is also possible; there is recent experimental evidence of this in molecules.

Tunnelling from multiple atomic energy levels is modelled and a subtle shift in ionization probability is expected as a function of intensity. We have measured focal-volume-dependent ionization of xenon using NIR ultrashort pulses generated at Artemis. As the experiment is able to resolve ionization as a function of intensity, the ionization probabilities can be extracted and compared directly to theory.



Predicted ionization probabilities of xenon exposed to a few-cycle laser pulse. Ground-state only ionization (dashed line) and ionization from all 5p and 5s states (solid line) are compared; we anticipate the intensity offset will be experimentally observable.

Ultrafast dynamics of electronic structure in complex materials



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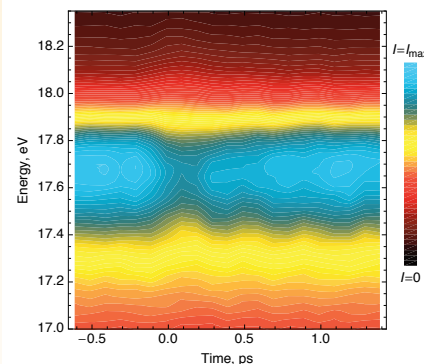
H. Berger (EPFL, Lausanne),

S. Kaiser, (University of Hamburg, Germany)

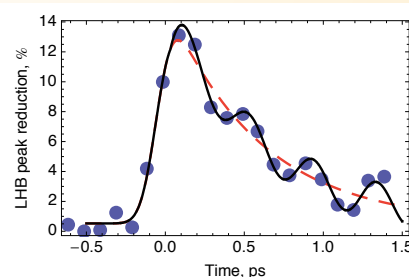
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We have used the new materials-science beamline at Artemis to carry out time-resolved photoemission studies of the layered Charge-Density-Wave and Mott-insulating compound 1T-TaS₂. We drove the system into a transient metallic phase using ultrashort pulses of laser light, and used high-harmonic UV pulses to generate photoelectrons, revealing the changes in electronic structure. After photoexcitation there is a collapse in intensity of the Lower Hubbard Band, transfer of spectral weight towards the Fermi level, and subsequent oscillation of the position of the band edge at the CDW amplitude mode frequency. These features agree with those seen at $k=0$ by Perfetti et al. [PRL **97** 067402 (2006)]. Our angle-integrated measurements additionally show an oscillation in the LHB peak intensity, a new effect likely due to the influence of the CDW amplitude mode on the band structure near $k=\pi/a$. Further experiments will study the detailed dispersion of the dynamical response.



Photoelectron intensity map of response to photoexcitation.



Reduction in LHB intensity. Solid line: Full fit. Dashed line: Exponential component only.

Ultrafast time-resolved photoelectron imaging of excited state molecular dynamics: S₂-S₁ internal conversion in the DABCO molecule



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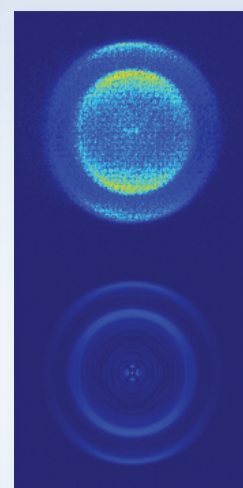
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Femtosecond time-resolved photoelectron imaging was applied to the ultrafast internal conversion dynamics in photoexcited 1,4 diazabicyclo[2.2.2]octane. A systematic investigation of the electronic relaxation dynamics upon the vibrational energy of the molecule was undertaken by scanning the pump wavelength between 236.1-251.1 nm. The temporal evolution of the photoelectron spectrum and angular distribution was measured. Analysis of the data is ongoing.



Pump pulse at 236.1 nm and probe pulse at 390 nm. The raw experimental data is shown at the top, and the Abel inverted data is shown below. The outer ring corresponds to photoelectrons from ionization of the S₂ electronic state, and the inner ring corresponds to ionization of the S₁ electronic state.

Biology

Probing metal complex dissociation in cells by fluorescence microscopy



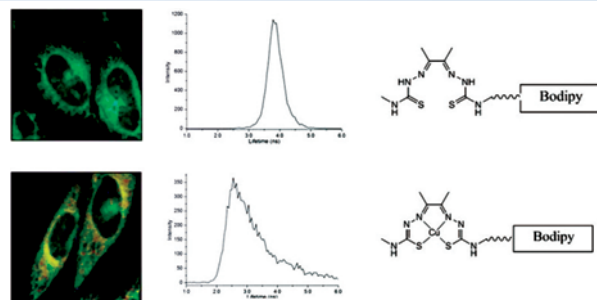
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Metal complexes of thiosemicarbazone species are known to be important biological agents, and have received considerable interest in terms of radio-imaging and therapy; however, to date the precise mechanism and cellular localisation of these drugs is uncertain. Whilst PET (Positron Emission Tomography) and SPECT (Single Photon Emission Computed Tomography) imaging can provide invaluable in vivo diagnostic information at the mm level, often little is known about the fate of the metallic radionuclides of these complexes once they enter cells. Fluorescence imaging has resolution which is currently in the order of 0.1-10 μm and to this end we have attached fluorophores to the thiosemicarbazone structures to aid



Emission lifetime maps for free bodipy/ATSM ligand(1) (top) and copper complex(3) (bottom).

elucidation of the mechanism of uptake and localisation of these metal labelled conjugates. One and two photon excitation lifetime measurements and images were determined on a series of thiosemicarbazone derivatives bearing potent inherent fluorophores. Findings indicate that careful choice of fluorophore is necessary to avoid influencing the behaviour of the complexes dramatically.

Biocompatible microstructured materials for tissue engineering



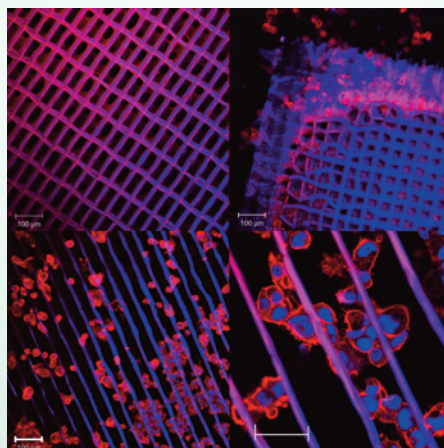
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In this project microstructuring of biocompatible materials was investigated via two-photon polymerisation (2PP), a laser-based direct write technique.

In 2PP a femtosecond laser is used to initiate the polymerisation via two-photon processes (2 near-IR photons are absorbed by an UV-sensitive photoinitiator). This results in a highly localised polymerisation at the focal point, and allows for 3D direct writing with excellent feature resolution (sub-micrometer). This process has been investigated for the production of biocompatible and biodegradable scaffolds for tissue engineering, using photocurable polylactide-based materials. In this project millimetre-sized patterns were written with micrometer resolution. The scaffold material showed good biocompatibility and distinct differences in neural cell growth were observed on these scaffolds depending on the scaffold geometry.

NG108-15 Cells on Microstructured Surfaces after 48h in culture. Cells have attached successfully to parallel lines of Microstructured PLA (Polylactic Acid) however they did not attach to square or rectangular structures indicating a degree of surface selectivity. Scale bars top and left, 100 μm , bottom right 10 μm .



The plant secretoryome: protein-protein interactions in the higher plant secretory pathway



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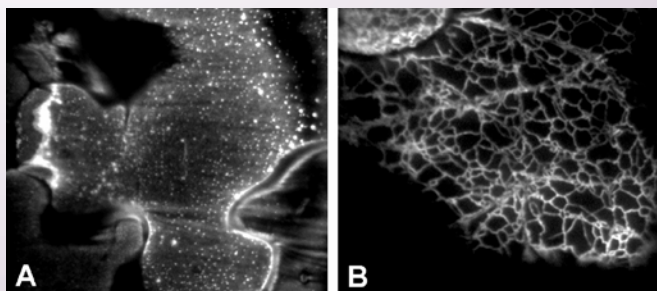
We have used FRET/FLIM to investigate protein interactions in the endomembrane system of plant leaf epidermal cells, TIRF microscopy to study the endomembrane system adjacent to the plasma membrane and are developing an optical tweezer set up on a TIRF microscope.

A family of endoplasmic reticulum proteins are involved in inducing curvature of the membranes of the plant endoplasmic reticulum (ER). As part of a project on the function of these proteins FRET/FLIM using fluorescent protein constructs in living

tobacco leaf cells was used to show that in the ER membrane the reticulin proteins can interact with each other forming hetero-and/or homo-oligomers [Sparkes et al. Plant Cell (2010) 22, 1333].

FRET/FLIM has also been used to study putative interactions between Golgi matrix proteins and small regulatory GTPases. Whilst TIRF imaging has successfully imaged ER, Golgi and the distribution of plasma membrane proteins at high resolution in tobacco and arabidopsis leaf epidermal cells.

A. TIRF image of the plasma membrane of a tobacco leaf epidermal cell expressing GFP-LTI6B showing punctae and a filament associated with the PM (image courtesy of A. Martinieri).
B. TIRF image showing cortical endoplasmic reticulum in a similar cell labelled with GFP-HDEL.



Supra-molecular rules in signalling networks: A single molecule comparative study in cells and tissues



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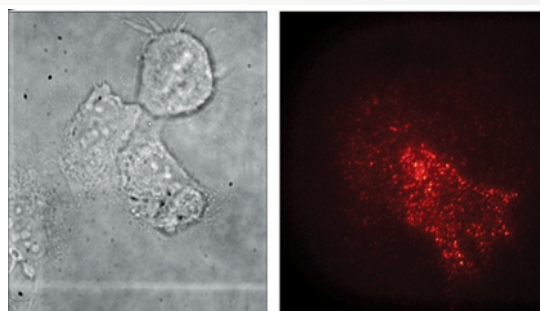
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We have developed single molecule and other fluorescence techniques on the Octopus facility, combined with simulation, modelling and analysis techniques as part of a programme in the Lasers for Science Facility (LSF). Our first aim is to define patterns of conformation association, activation and signal transduction for the 4 members of the ErbB family and their correlation with plasma membrane proximal receptor traffic and signalling using cultured cells. We also aim at deriving models of ligand-induced

behaviour using single molecule and ensemble FLIM and systems predictions and to establish how this is influenced by feed-back loops and perturbations. By testing these models on primary epithelial cells and tissues we will adapt the models iteratively to create a framework of response prediction for the network. Our ultimate goal is to derive probes and/or algorithms based on the models, suitable use with fixed or fresh human tissues to determine model robustness and network predictability in this real world setting.

Wide field transmission (left) and single molecule fluorescence (right) images of HeLa cells. The EGFR molecules at the cell surface were labelled with EGF tagged with the red emitting fluorophore Atto647. The spots on the figure on the right show the location of single EGF/EGFR complexes in different oligomerisation stages. The number of fluorophores is determined by the number of photobleaching steps in each spot.



The role of BER proteins in the repair of DNA damage induced following NIR multiphoton laser microbeam irradiation



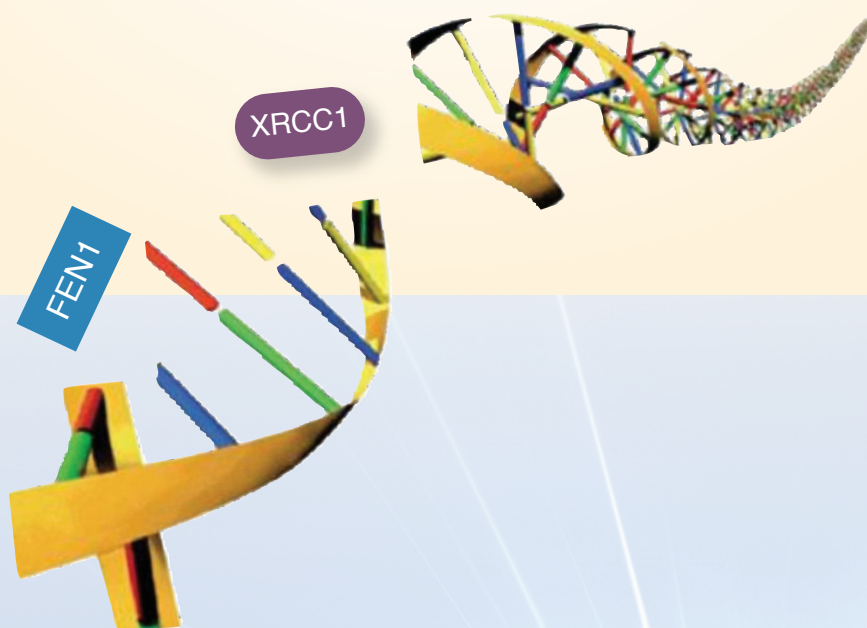
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DNA damage induced by radiation has been shown to increase genome instability and mutations, which could in turn lead to the formation of cancerous cells. Ionising radiation may be considered as a two-edged sword; it may induce cancer or other adverse responses in normal tissue or alternatively it may lead to killing of tumour cells, of considerable importance in radiotherapy and cancer treatment. The repair of ionising radiation induced DNA damage has therefore been extensively studied in mammalian systems as it is essential to maintain genome stability.

The use of NIR laser microbeam has demonstrated its feasibility to study the induction of lesions in genomic DNA in living cells. Key proteins involved in a repair process required to maintain genome stability, base excision repair, are recruited and lost from sites of induced DNA damage at different speeds possibly reflecting the repair of different types of DNA damage. The identification of these types of damage which are difficult to repair may advance our understanding of cancer induction.



Chemistry

Time resolved infrared analysis of AppABLUF using isotopic labeling at mutant studies



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AppA is the first and best characterized BLUF domain photoreceptor and is found in Rhodobacter sphaeroides where it acts as an antirepressor of photosystem biosynthesis. Our studies have focused on the N-terminal domain of AppA responsible for sensing light. Previous studies determined the formation of a signaling state which is characterized by a 10 nm red shift in λ_{max} of the flavin chromophore. The goal of the current study is to identify differences in the chemical structures of

the dark and signaling state of AppA. Both AppA's dark and light excited state dynamics have been studied using time resolved infrared spectroscopy (TRIR). We have also used isotope labeling of both the flavin chromophore and of the protein in order to identify major differences in the TRIR spectra of light AppA and dark AppA. Additionally, we have studied structurally conservative AppA mutants which play a crucial role in photoactivity.

High-resolution stimulated Raman spectroscopy with photoacoustic detection (PARS) of formic acid dimer



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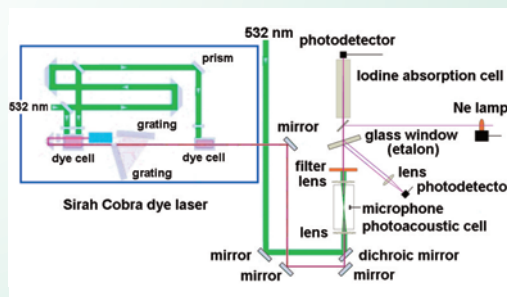
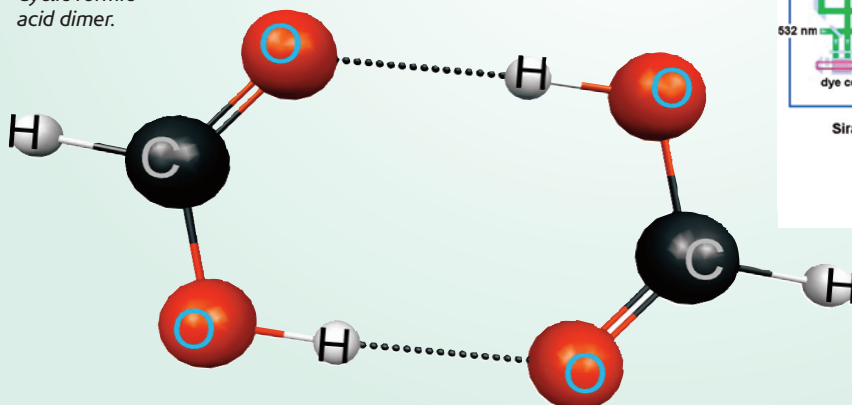
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A stimulated laser Raman experiment with optophone photoacoustic detection has been set up which allows high-resolution Raman spectroscopy of gas-phase species in hostile environments, PARS. With this technique, we have observed the CH-stretching vibration of formic acid dimer which shows a distinct shift towards higher wavenumber ('blue shift'). Such shifts have in the past been attributed as a typical sign of intermolecular bonding. In the cyclic dimer, however, CH is not involved in hydrogen bonding. We propose that this is

a secondary effect where increase of electron density to the C=O bond by hydrogen bonding with OH has an electron withdrawing effect. This increases the polarity and strength of the C-H bond, thus inducing a shift towards higher wavenumber.

In future work, PARS can also be used to observe the OH-stretching vibration of cyclic formic acid dimer to study the concerted double-proton transfer in the two equivalent OH...O hydrogen bonds.

Cyclic formic acid dimer.



Scheme of the PARS setup with wavelength calibration.

Transient 2D-IR spectroscopy of [FeFe]hydrogenase enzyme model compounds

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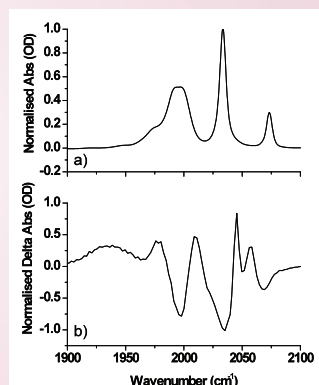
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Ultrafast multidimensional infrared spectroscopy is a useful tool for probing complex reacting systems due to the ability to spread the molecular response over two frequency dimensions and thus give access to previously inaccessible information. Furthermore, the inherent ultrafast time resolution of these techniques provides the ability to observe reactions in real time. Here, transient 2D-IR (T-2D-IR) spectroscopy has been employed to determine the infrared absorption frequencies and coupling patterns of the photoproduct vibrational modes of the species formed following photolysis of $(\mu\text{S}(\text{CH}_2)_3\text{S})\text{Fe}_2(\text{CO})_6$, a model compound of the [FeFe]hydrogenase enzyme active site, in cyanoheptane solution. Double difference T-2D-IR analysis has revealed a number of peaks due to a single

photoproduct and they show spectroscopy consistent with the formation of a pentacarbonyl solvent adduct species.



a) FTIR spectrum of $\sim 1\text{ mM}$ solution of 1 in cyanoheptane solution. b) TRIR spectrum of the same solution following photolysis at 350 nm. The data shown corresponds to a $\text{UV}_{\text{pump}} - \text{IR}_{\text{probe}}$ delay time of 200 ps.

Ultrafast TRIR studies on tetraazidosilicon complexes

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Photoinitiated CO-release molecules

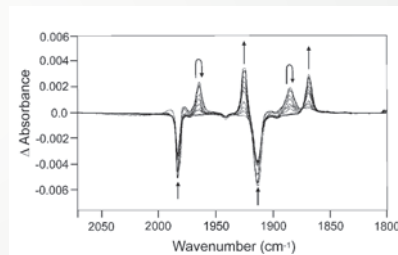
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Carbon monoxide (CO) is a toxic odourless gas. Surprisingly however it is generated in living organisms during the degradation of heme by the enzyme heme oxygenase. In low concentrations carbon monoxide gas acts to dilate blood vessels in a manner similar to nitric oxide and it also has some anti-inflammatory and anti-apoptotic properties. We are currently investigating metal carbonyl complexes as possible CO Releasing Molecules (CORM) for use as therapeutic agents. Fundamental to the design of new CORM is an understanding of the photophysical processes leading to CO-loss. Two metal carbonyl systems have been chosen, $(\eta^6\text{-arene})\text{Cr}(\text{CO})_3$ and $(\mu\text{-alkyne})\text{Co}_2(\text{CO})_6$. Photoinduced CO loss from $(\eta^6\text{-arene})\text{Cr}(\text{CO})_3$ occurs from a vibrationally cold excited state while

photolysis of $(\mu\text{-2 pyreneacetylene})\text{Co}_2(\text{CO})_6$ results in CO loss producing $(\mu\text{-2 pyreneacetylene})\text{Co}_2(\text{CO})_5(\text{S})$ ($\text{S} = n\text{-heptane}$). These results will assist in the design of new molecular systems with specific spectroscopic and photochemical properties ideal for use as CORM agents.



The TRIR spectra obtained following 400 nm excitation of $(\eta^6\text{-Benzene})\text{Cr}(\text{CO})_3$ in $n\text{-heptane}$, spectra were recorded at 1, 7, 12, 21, 51, 70, 106, 160, 350, 500, 750, 1000 ps after the excitation pulse.

Excited-state dynamics of adenine thymine dinucleotides: influence of stacking



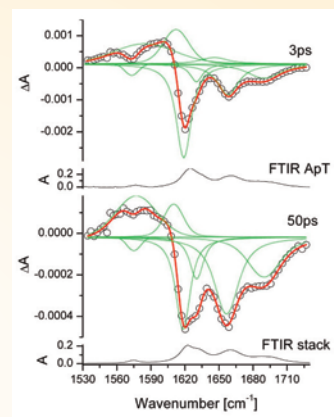
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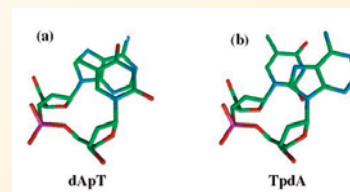
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Individual bases of DNA typically decay on the ultrafast timescale however in a sequence longer-lived species are observed which are susceptible to chemical change. In DNA bases interact through stacking and hydrogen bonding and understanding their influence on the excited states is important. We studied two simple sequences, dApT and TpdA, which are both stacked and unstacked in solution (38% and 20% stacked respectively). Using picosecond time-resolved infrared (ps-TRIR) spectroscopy we observed two transient species, a short-lived species (5 ps), of the unstacked form and a longer-lived species from the stacked form. The yield and the lifetime of the longer-lived species differed significantly in dApT (75 ps) and TpdA (50 ps). Importantly, its distinctive spectroscopic signature (1500-1600 cm^{-1}) is identical to that observed for the long-lived state in poly(dA-dT). The study demonstrates the influence of stacking and confirms that the excitation in the polymer is localized on the dinucleotide unit.



ps-TRIR spectra of 10 mM dApT following UV excitation (300 fs, 267 nm) upper Voigt fitted data at 3 ps and ground state FTIR spectrum for dApT lower Voigt fitted data at 50 ps and ground state FTIR spectrum of stacked component of dApT



Overlap in the stacked form of (a) dApT and (b) 5'-TpdA-3'. Molecules were drawn using the ChemSW® package.

Antigens laid bare: the intrinsic conformation of Lewis^x



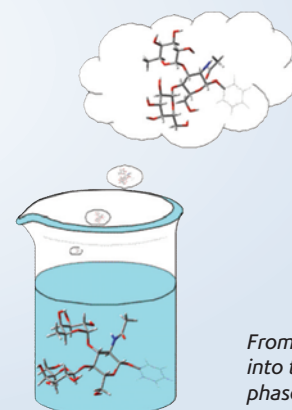
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The trisaccharide, Lewis^x is expressed by many cancers of epithelial origin and is an important immunotherapeutic target. Knowing its structural preference(s), particularly in its bioactive conformation, is a key requirement in designing drugs which might inhibit its binding and activity. In aqueous or protein bound environments Lewis^x adopts relatively rigid, compact structures with its terminal units, galactose and fucose, stacked in parallel. But is this its intrinsic conformation or one controlled by the environment? A combination of laser desorption and tunable infrared laser techniques, which can interrogate individual molecular conformers isolated in the rarified environment of a cold, molecular beam, and quantum chemical computation has provided the answer.

The intrinsic structure is not stacked but hydrogen bonded and the results of these experiments suggest that the environment, particularly water, plays a key structural role in controlling the conformation of Lewis^x and in consequence, its biological function.



From solution into the gas phase.

Carbohydrate molecular recognition: probing CH- π interactions



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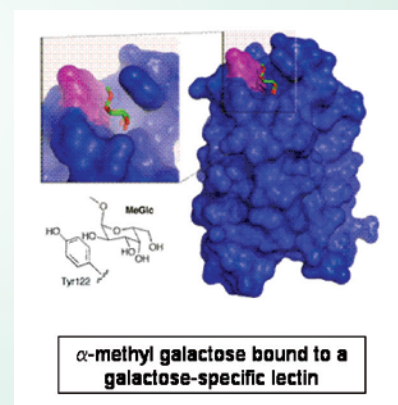
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Interactions between carbohydrates and proteins are intimately involved in the biological machinery of a vast number of biological processes: mammalian infection, inflammation, fertilization and cellular recognition for example. Molecular recognition by specific carbohydrate binding modules is controlled by the disposition, conformation and configuration of the locally interacting sites. Fine controls are often provided by hydrophobic interactions between the apolar regions of the carbohydrate and aromatic residues, thought to be promoted (in aqueous environments) by the exclusion of surrounding water molecules from the apolar interface though binding to aromatic residues could also involve dispersive, so-called "CH- π " interactions. These have now been identified in model carbohydrate-arene complexes, isolated in the gas phase free from neighbouring

solvent molecules and stabilized at low temperatures in a molecular beam. The molecular complexes, probed through infrared laser spectroscopy, adopt stacked structures supported solely by dispersive "CH- π " interactions between the pyranose and aromatic rings.



Stacking in a carbohydrate-protein complex.

Polymer coil-globule transition dynamics on the nanosecond to second time scale



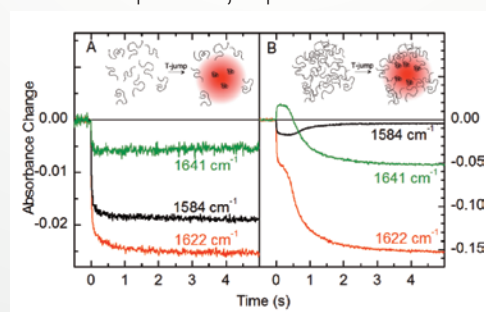
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When heated over the lower critical solution temperature (LCST), poly(N-isopropylacrylamide) undergoes a change in conformation from a random coil to a collapsed structure which then leads to phase separation. This effect is investigated for a wide range of applications, including drug or DNA delivery and control of enzymatic activity. Although the transition has been studied extensively, its dynamics has received only little attention so far. We have used laser-induced temperature jumps and IR

spectroscopy to follow the polymer collapse on time scales ranging from a few nanoseconds to several seconds. An increase of the temperature over the LCST leads to a local collapse of polymer sections within nanoseconds due to cooperative hydration, followed by much slower phase separation. Partial phase separation on the 100 ms time scale could be observed, as shown in the Figure, but only at concentrations resulting in significant overlap of the polymer chains.



IR absorbance changes after a temperature jump over the LCST at polymer concentrations of 10 mg/ml (A) and 45 mg/ml (B). Whereas the signal in (A) is dominated by heating of the solvent (water), (B) shows a decrease of the polymer absorbance, indicating that polymer is removed from the heated sample volume (phase separation) when polymer chains overlap.

Physics

Using tip enhanced femtosecond lasers to create graphite nanostructures on diamond



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(ISIS, STFC, Rutherford Appleton Laboratory, UK)

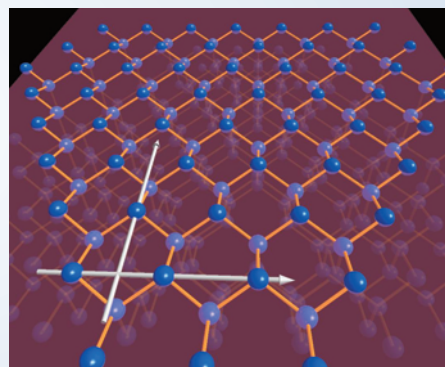
R.B. Jackmann
(University College London, UK)

S. Bennington,
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Graphene is the new wonder material; it is a zero band gap semiconductor and has electron mobilities two orders of magnitude greater than in silicon. Our interest is in thin ribbons of graphene with well defined edge states predicted to be highly magneto-resistive and potentially even half-metallic, the holy grail of spintronics research.

From our previous work we knew that a femtosecond laser will convert the (111) surface of diamond into thin layers of graphite, the interaction occurring so quickly that the bonds reform before there is any significant heating. In this work we used the laser in conjunction with an atomic force microscope to enable us to produce structures smaller than the wavelength of light.

Although we made sub-diffraction limit structures on silicon with relative ease, in these initial experiments we were unable to make structures on the non-conducting surface of the diamond.



The (111) surface of diamond showing the puckered graphite like arrangement of the atoms. The two arrows show the armchair edge direction ($2\bar{1}\bar{1}$) or the zig-zag edge ($0\bar{1}\bar{1}$).

Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing



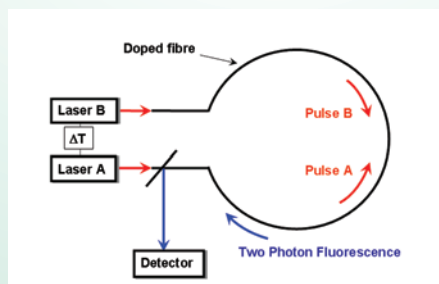
I.S. Ruddock, C.J. Dalzell, T.P.J. Han
(University of Strathclyde, UK)

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Distributed fibre sensing based on the temperature or strain dependence of the fluorescence decay time or spectrum can only be achieved using two-photon excitation. An enhanced fluorescence signal will be produced where two counter-propagating excitation pulses overlap in a doped fibre if the sum of their photon energies excites the upper level of the ions. The Laser Loan Pool NS3L optical

parametric oscillator was used to investigate the nonlinear spectroscopy of glass and crystal doped with various lanthanide and transition metal ions. The two-photon absorption cross-section was obtained by comparing the two-photon excited fluorescence (TPF) yield with that for single-photon excitation for the same transition under identical conditions. Related experiments on TPF in doped glass and fibre are outlined in the accompanying synopsis, "Distributed sensing by time-correlated two-photon excited fluorescence in rare earth doped optical fibres" by I S Ruddock et al..

Schematic diagram of a distributed sensor based on time-correlated TPF in a doped optical fibre. Counter-propagating pulses (A) and (B) meet at a location depending on their relative time delay, ΔT . The TPF generated at their overlap is guided along the fibre and detected.



Distributed sensing by time-correlated two-photon excited fluorescence in rare earth doped optical fibres



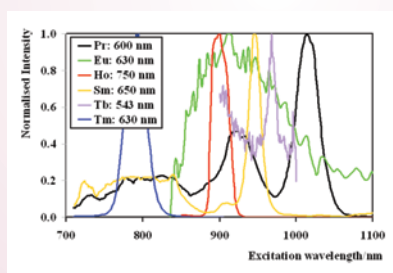
I.S. Ruddock, C.J. Dalzell, T.P.J. Han
(University of Strathclyde, UK)

D.B. Hollis
(University of the West of Scotland, UK)

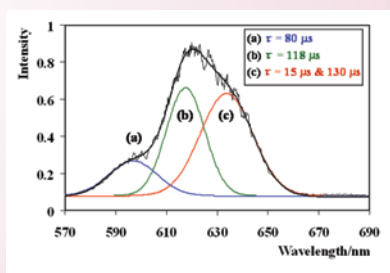
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Potential dopants for distributed fibre sensing based on two-photon excited fluorescence (TPF) were identified by single- and two-photon excitation spectroscopy of bulk samples of rare earth doped glass. Using the tuneable near-infrared idler and the visible signal outputs of the Laser Loan Pool's NSL3 optical parametric oscillator, two-photon and two-step excited fluorescence was observed in praseodymium, samarium, europium, gadolinium, terbium, dysprosium, holmium and thulium. From this list, praseodymium was selected for detailed investigation in single-mode fibre geometry on account of

its convenient excitation and TPF wavelengths and its potentially negligible fluorescence self-absorption due to the transitions of interest near 600 nm terminating above the ground state. An outline of fluorescence based distributed fibre sensing and related experiments on determining the two-photon absorption cross-section are given in the accompanying synopsis – “Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing” by I. S. Ruddock et al..



Two-photon and two-step excited excitation spectra for a range of rare earth doped glasses of different composition. The corresponding emission wavelengths are indicated in the legend. For praseodymium (black line), note the TPF excitation peak at ~930 nm and the adjacent two-step excited fluorescence peak at ~1014 nm.



The TPF spectrum for praseodymium doped silica fibre showing the composite emission line at ~600 nm, the de-convolved components and their respective room temperature decay times, τ .

Artemis

Monochromatised XUV beamline for ultrafast time-resolved ARPES



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W.A. Bryan, G.R.A.J. Nemeth
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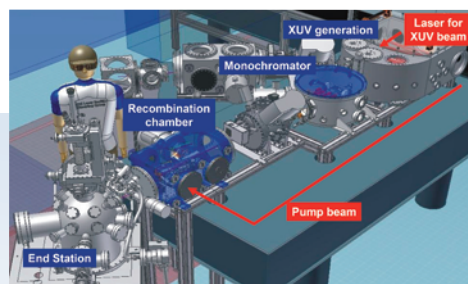
A. Cavalleri, J. Petersen, N. Dean
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S. Dhesi
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The short pulses of coherent XUV radiation produced through high harmonic generation have enabled the study of ultrafast electron dynamics in atomic systems and simple molecules. The Artemis facility now aims to exploit these XUV pulses to investigate ultrafast dynamics in experiments spanning gas-phase chemistry of polyatomic molecules and condensed-matter physics of complex materials.

We have built an XUV beamline that delivers short pulses of monochromatised XUV, produced through high harmonic generation, to an end-station optimized for photo-emission experiments on condensed matter.



Layout of the XUV beamline including the Harmonic generation chamber, the monochromator chamber to select the photon energy, the recombination chamber for pump-probe experiment and the experimental chamber for time resolved ARPES.

A specially designed monochromator allows us to select a single harmonic and preserve also the short pulse duration (10 - 30 fs). We have shown that the beamline can achieve 250 meV energy resolution and 30 fs temporal resolution, enabling us to carry out first measurements of angle- time- resolved photoemission with XUV pulses.

Spin and angle resolved photoemission with fs laser source: calibration of spin detector



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B. Ressel, and F. Parmigiani
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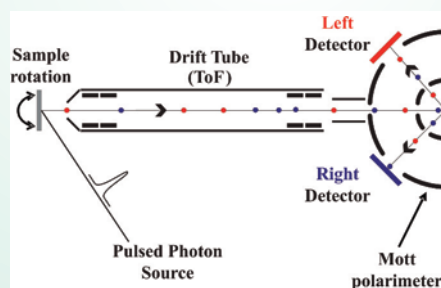
E.A. Seddon (Manchester University, UK)

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Angle resolved photoemission is a very powerful technique to explore the electronic structure of materials. Over the past three decades this spectroscopic technique has well matured partly due to the development of intense synchrotron radiation and bright fs-laser sources. Measuring the spin as well as the emission angle of the photoelectron is particularly interesting to study the magnetic properties of ferromagnetic systems or

systems with large spin-orbit coupling [1-2]. The intrinsic low efficiency of the electron spin detection makes these experiments very challenging. This is why, using a laser source with a novel electron analyzer (electron time-of-flight) is a real step forward for this type of experiment particularly in terms of acquisition time and resolution. Furthermore by combining the ToF-Spin analyzer to the Artemis facility, one can study the electron spin dynamic in the sub 10-fs time domain which is of great interest to explore the switching mechanisms in ferromagnetic systems for spintronic devices.

- 1 J.-H. Park et al., Nature, **392**, 794 (1998).
- 2 K. Sakamoto et al., Phys. Rev. Lett., **102**, 096805 (2009)



Top view of the experimental setup in normal emission geometry. The photoelectrons travel through the drift tube up to the Mott

polarimeter where they are spin resolved. The arrival time of the electrons at each detector is measured and converted to kinetic energy.

Few-cycle carrier-envelope-phase controlled laser pulses for time resolved science at the Artemis facility



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R.B. King
(Queen's University Belfast, UK)

C.M. Cacho, I.C.E. Turcu and E. Springate
(CLF, STFC Rutherford Appleton Laboratory, UK)

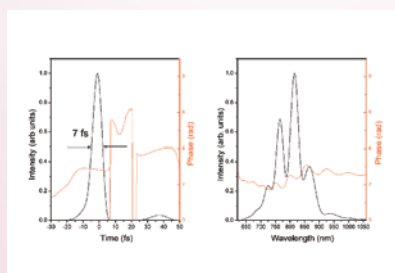
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When the duration of a laser pulse reaches the few-cycle level, the phase of the laser electric field relative to the pulse envelope (the carrier-envelope phase, CEP) can have a significant effect on the physics of the interaction. Examples of this are the generation of isolated attosecond pulses [1] and the control of electron localisation during molecular dissociation [2], both of which require few-cycle pulses with stable CEP.

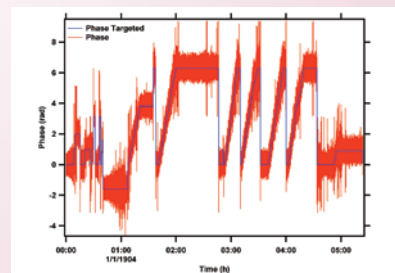
The Artemis facility is constructed around a CEP stabilised laser system with an output of 780 nm, 30 fs, 14 mJ pulse at 1 kHz. Part

of the energy can be focused into a hollow fibre to generate few-cycle pulses. Recent improvements have reduced the pulselength to 7 fs, with 0.5 mJ per pulse. The long-term stability of the CEP stabilisation is 325 mrad rms for over five hours.

1. A. Baltuska et al, " Attosecond control of electronic processes by intense light fields", Nature 421, 611 (2003).
2. M. F. Kling et al, "Control of electron localization in molecular dissociation", Science 312 246 (2006).



FROG measurement of 7 fs, 0.5 mJ pulses produced in the hollow fibre system showing retrieved temporal intensity and phase. The FROG error was 0.006.



CEP control over five hours with 2 mJ/pulse and 325 mrad rms stability. Blue line: programmed target phase. Red line: measured phase.

Astra

Vibration in Gemini and engineering modifications (interim report)



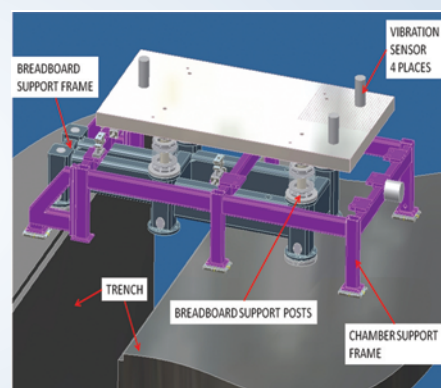
S. Blake, R. Pattathil, P.S. Foster
(CLF, STFC, Rutherford Appleton Laboratory, UK)

H. Huang
(Diamond, UK)

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Intermittent ground borne vibration delayed and plagued early experiments in Astra Gemini. The vibration manifested itself in an elliptical or figure of eight movement of the spot at the interaction point. After analysis the source was traced back to the main magnet power supply of the neighbouring ISIS facility. The main source at 50 Hz was amplified by the support structure, multi component breadboard and key optical mounts increasing the amplitude of the vibration at each stage. Large structural changes were carried out to the main optics table and associated framework, parabola chamber and the key optical mounts. The initial testing post upgrade is positive. The image on the right shows the main optics table (breadboard), supporting structure which spans the trench leading to ISIS and the change to the key optical mounts.

The full report details the vibration scan results, changes to the hardware and initial conclusions.



The new structure and the location of the four sensors.

Temporal and spatial overlap monitoring of the dual-beam layout in Astra-Gemini TA3

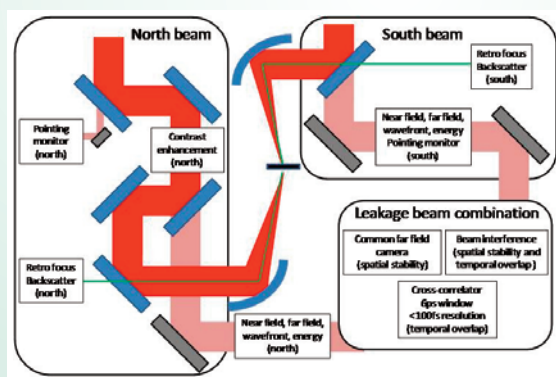


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P. P. Rajeev, D. Neely
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The Astra-Gemini target area is now up and running for dual beam experimental operations. It is now essential to have spatial and temporal overlap of the two beams at the target position. Two extra diagnostics have been added to the target area to monitor these overlaps on a shot-to-shot basis.

To monitor the spatial overlap, leakage from each of the beams is taken, then combined, focussed, and split to a near-field and a far-field camera. The near-field will be used to indicate the temporal overlap of the two beams and the jitter in the spatial alignment. The far-field will monitor the spatial stability of the overlap of the two beams.



A cross-correlator diagnostic has been installed in order to monitor the temporal overlap. Leakage is taken from each of the beams which are both split and overlapped with two BBO crystals to generate two cross correlation traces, one with a time window of ~ 6 ps and one with a > 6 ps window.

Schematic diagram of the beam arrangements in Astra-Gemini TA3. Red indicates the main beamlines and pink indicates leakage beams.

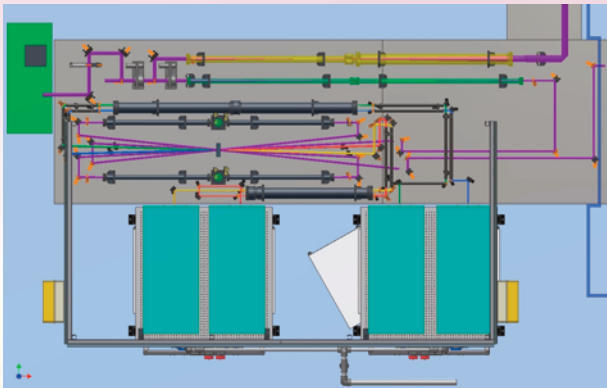
Upgrade of Astra amplifier 3 and the Astra interlock system



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C. John, B. Landowski, A. Tylee
(CLF, STFC, Rutherford Appleton Laboratory, UK)

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Last year a significant upgrade was carried out to the old Astra amplifier 3, which has improved its performance and removed one of the least reliable commercial lasers from the system. The old Macholite laser was replaced with four Quanta-Ray PRO-350 Nd:YAG lasers, and a new water cooling system installed. The layout of the amplifier was redesigned for better accessibility: before the changes, the beam expansion tubes were on a raised framework above the amplifier, but now everything is on the same level. The new layout is shown in the diagram. The performance of the amplifier is better than before, with a typical output of 1.2 to 1.3



Schematic of the new layout for Astra Amplifier 3.

Joules at 800 nm. During the same period, the old PC-based interlocks were replaced with an up-to-date system based on PLC technology, giving improved safety and full compatibility with other areas of the CLF.

Improved post-experiment data analysis at Astra Gemini



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We have further improved the gathering and analysis of diagnostic data over the course of an experiment. In conjunction with the Principal Investigator, we identify the laser parameters (singular values, trace images and camera images) which are key to that experiment and make them available to users as Comma-Separated Values (CSV) files. This CSV data can be downloaded and incorporated into analysis programs written for target area diagnostics. Singular value data streams are subsequently averaged and displayed;

traces are averaged and displayed as a Scalable Vector Graphics (SVG) images. The laser operator can subsequently view this data and, for image data streams such as far-field profiles and FROG traces, select the image which represents a typical image for that day. In this way we compile an overall summary table detailing the average trace plots, representative images and a scatter plot of X/Y alignment pairs on a shot-by-shot, day-by-day basis.

date	#shots	s comp ff	s comp spec trace	s uncomp spec trace	s frog master image	s comp ff image	operator's comments
2010-02-14	35						Positioning good.
2010-02-15	81						
2010-02-16	41						Vibration problems?

Three day's worth of representative images and plots.

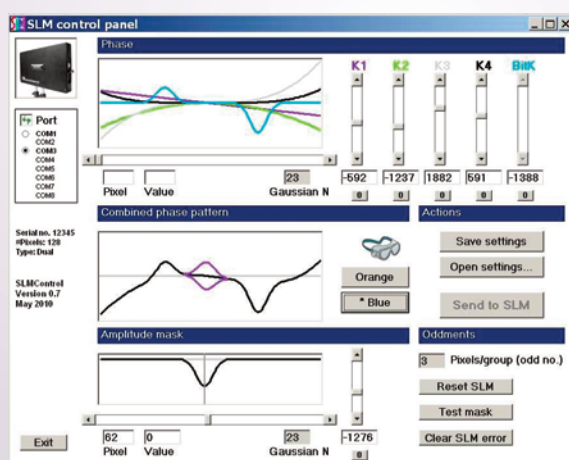
A new phase (and amplitude) for Astra Gemini: using a spatial light modulator



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We have developed a new method for correcting the spectral phase and amplitude of the Astra Gemini laser, based on a spatial light modulator (SLM). This device uses voltages applied to a pair of liquid-crystal arrays to modify different parts of the spectrum in a spatially-dispersed laser beam.



The sum of the voltages controls the phase and the difference controls the spectral amplitude. The existing phase corrector (Dazzler) suffers from crosstalk between phase and amplitude corrections: the advantage of the SLM is that such crosstalk is minimised. The SLM is positioned at the mid plane of a 4F zero-dispersion optical setup using blazed 600 groove/mm gratings. It is controlled through a custom interface that allows the user to set the amplitudes of four polynomial phase terms and to apply localised phase and amplitude adjustments. The interface is designed for use by operators wearing laser protective eyewear.

View of the SLM control system user interface.

Ultrafast gated imaging of laser produced plasmas using the optical Kerr effect



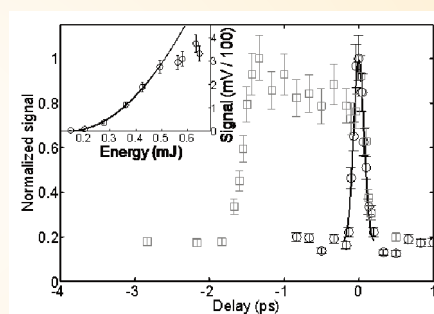
D.R. Symes, U. Wegner, H.-C. Ahlswede,
M.J.V. Streeter, P.L. Gallegos, E.J. Divall,
P.P. Rajeev, D. Neely (CLF, STFC Rutherford
Appleton Laboratory, UK)

R.A. Smith
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Optical imaging provides a versatile diagnostic for investigations of plasmas generated under intense laser irradiation. Electro-optic gating techniques operating on the >100 ps timescale are commonly used to reduce the amount of light detected from self-emission of hot plasma or improve the temporal resolution of the detector. The use of an optical Kerr gate enables a superior dynamic range and temporal resolution compared to electronically gated devices. We have applied this method using the hollow fibre pulse compressor in Astra TA2 to demonstrate enhanced imaging of laser produced plasmas. Employing fused silica, which has an ultrafast response, as the gating material we achieved a gate time ~ 100 fs. We also discuss alternative materials. We used the system to image plasma produced with moderate intensity irradiation and show the elimination of plasma self-emission from the image.

We conclude by exploring the possibility to produce a sub-10fs, high dynamic range "all optical" streak camera.



We use the optical Kerr effect to perform ultrafast gated plasma imaging. The opening time of the gate is ~ 100 fs in fused silica (black) compared to slower response materials. Shown by the throughput of the device versus delay between gating and probe beams.

Lasers for Science Facility

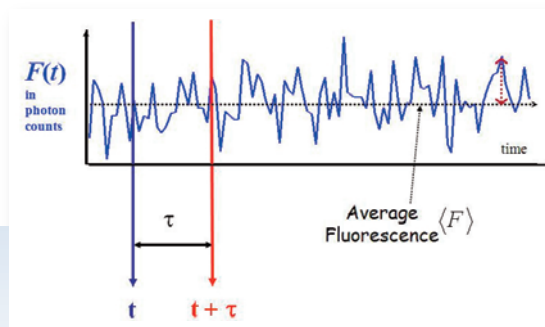
Two-photon fluorescence correlation spectroscopy: FCS



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Fluorescence correlation spectroscopy (FCS) is a powerful single-molecule detection technique used to characterise the dynamics and interactions of fluorescent species by recording and correlating their fluctuations in fluorescence intensity within a microscopic detection. To demonstrate the potential of



Calculating the autocorrelation function.

this technique using our TiSa confocal setup, the diffusion coefficient of the Rhodamine dye was measured for different concentrations and laser powers. Surface diffusion has been recorded in vivo.

The ULTRA time-resolved IR, 2D-IR and T-2D-IR station



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Developments within the EPSRC Laser Loan Pool



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The Octopus imaging cluster: A new imaging facility in the Research Complex at Harwell

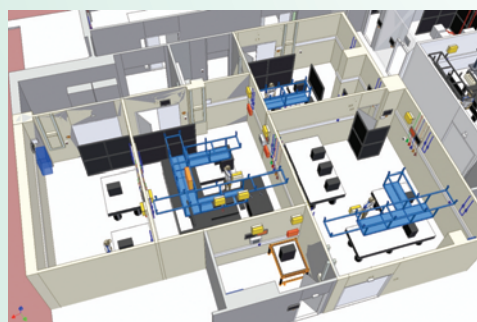


S.W. Botchway, D.T. Clarke, M. Hirsch,
A. Mackenzie, M.L. Martin-Fernandez,
S.R. Needham, S.K. Roberts, D.J. Rolfe,
A. Tylee, C.J. Tynan, S.E.D. Webb
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The past year has again seen major changes in the Lasers for Science Facility. The LSF's operations in the biological imaging area were relocated to the Research Complex at Harwell (RCaH) in April 2010. The Octopus cluster is a large scale imaging facility offered by the Functional Biosystems Imaging Group of the LSF which begun user operations and collaborative work in July 2010. Octopus is a new concept in multi-modal imaging. Currently in Phase I of its development, the

Octopus cluster has at its core a hub of 14 lasers (3 Ti:Sa, 2 OPOs, 3 supercontinuum and 6 CW). The laser sources provide excitation light to seven microscope systems, namely a total internal reflection fluorescence (TIRF) tweezers system, 3 single molecule fluorescence microscopes, each of which is specialised in a different single molecule imaging technique, two 1-photon confocal FLIM (Fluorescence Lifetime Imaging Microscopy) systems and a 2-photon confocal FLIM system laid out as shown left.



Layout of the Octopus cluster.

The central room is the laser hub with the different microscopes lab position around it. It also houses a two-photon confocal FLIM system. Bottom right: this room houses three systems, namely a total internal reflection fluorescence (TIRF) tweezers system, a one-photon confocal FLIM system and a

5-colour single molecule tracking system. Top right: This room houses a one-photon multicolour confocal FLIM system. Top middle: The control room (see Fig. 2). Top left: Storage room. Left: This room houses a 3-colour single molecule tracking system (top) and an 8-channel single molecule FRET and polarisation combined system (bottom). Bottom middle: Development room.

New data analysis software for time-resolved spectroscopy experiments in the Ultra laser facility

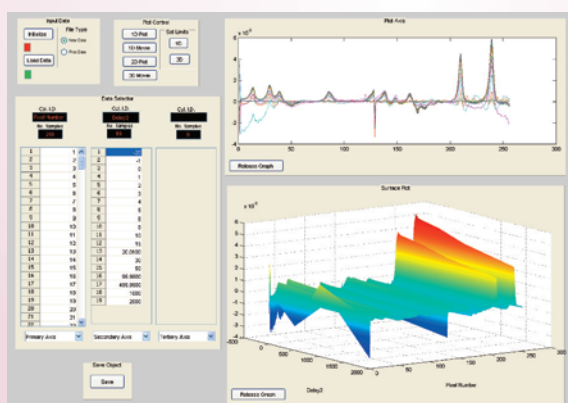


M.R. Pollard, G.M. Greetham, D.A. Robinson,
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New data analysis software has been developed in response to the start of operations in the Ultra laser facility and the significant increase in experimental data that it brings. The facility generates spectral data at a typical rate of > 50 spectra per experiment with ~ 15 experiments per day, which requires visiting scientists to perform quick and

efficient analysis of large data sets in order to guide their experiment. The need for expediency is reinforced because of the limited lifetime of precious biological and chemical samples that are potentially light or temperature sensitive. We describe the features of the software including correction for instrument variation (such as background intensity and detector-based electronics noise) and a flexible data display that allows visiting scientists to observe their data according to experimental parameters in two and three dimensions.



Screenshot of graphics display, showing data from sample of DMABN.

Active synchronisation of dual amplifier outputs for time-resolved spectroscopy

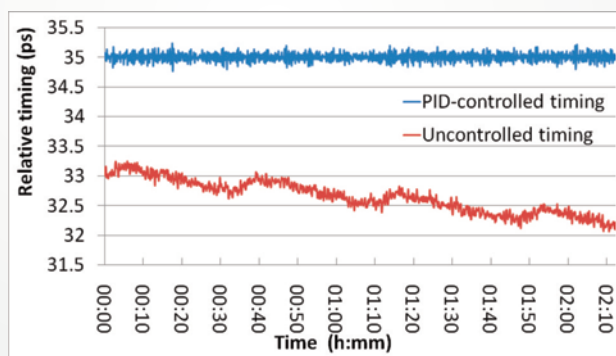


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Many of the time-resolved spectroscopy experiments in the Ultra laser facility take advantage of its unique dual synchronised 2 ps and 50 fs output pulses. The relative timing between the pulses is susceptible to thermal drift of potentially several mm that occurs in the optical paths within the laser amplifiers, which significantly affects experimental measurements.

We report the development of a control system that improves synchronisation of the two outputs to 40 fs rms, achieved using a scanning optical correlator system to measure the relative timing between the pulses and a feedback loop control system to ensure that the synchronisation of the two outputs is constrained to a defined set-point.



Comparison of relative timing over 2 hours.

Optical trapping of laser targets under vacuum for ion-beam production in the LIBRA programme



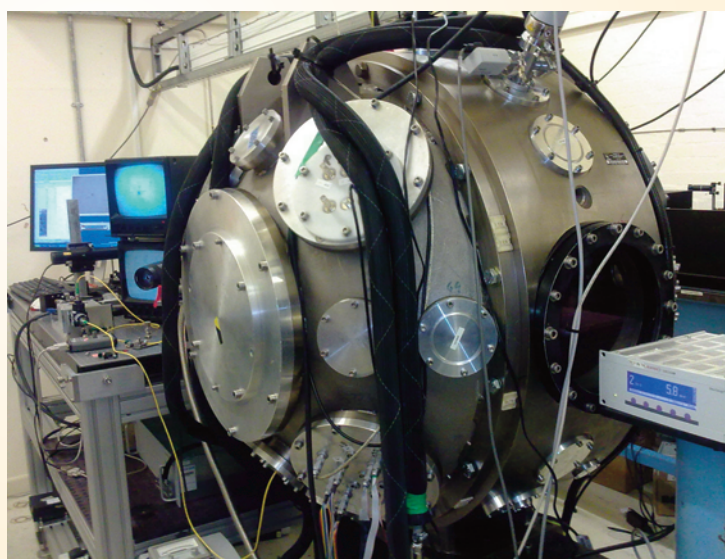
A.D. Ward, B. Li, T. Strange
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As part of the targetry package of the EPSRC funded LIBRA programme, we have successfully held a microscopic liquid droplet in the centre of a vacuum interaction chamber at a pressure of 5 mbar using laser-based optical levitation techniques. The droplet was held to a positional accuracy of 1 micron which demonstrates the concept is suitable for manipulating targets into a focused

high-power laser beam. In the report we outline the underpinning methodology and optical requirements for trapping micron-sized droplets using long-working distance, high numerical aperture objective lenses. By extension of this technology, our goal in the LIBRA programme is to hold solid targets for the purpose of generating ion-beams by laser irradiation.

The vacuum chamber and diagnostics in operation. Droplet stability, laser alignment and chamber pressure are continually monitored. Trapping can be achieved from 1000 mbar to 5 mbar before efflorescence of the salt droplet occurs (i.e. a crystal is formed). The droplet is observed to be stable in position to approximately 1 micron.



Vulcan

Close-in contrast measurements of the new ps OPCPA front end

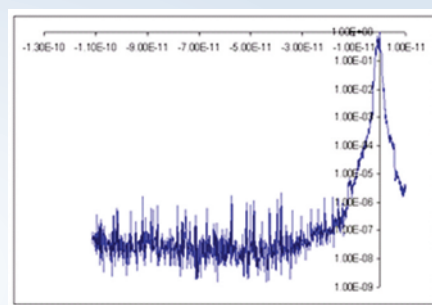


A.G. Boyle, I. Musgrave, M. Galimberti,
W. Shaikh, K. Lancaster, B. Costello,
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In high intensity laser matter interactions the contrast of the laser pulse can be crucial to the outcome of the physics. In this article we discuss the addition of a picoseconds optical parametric chirped pulse amplification (OPCPA) stage to the Vulcan front end improving the amplified stimulated emission.

The novel dual OPCPA pre-amplifier system is sufficiently stable to be injected into the rest of the Vulcan laser system and leads to a direct improvement of the ASE contrast by at least 2 orders of magnitude without degrading the performance of the laser system. The Vulcan Petawatt laser system now has an ASE contrast of 10^{10} at 1ns, at 100ps the contrast is better than 10^8 and the PF (Parametric Fluorescence) caused by the ps OPA starts at 30ps.



3rd order cross correlator (Sequoia) trace, which is a multi shot device that temporally scans the pulse.

Vulcan rod amplifier upgrade



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The Vulcan rod amplifier control system and power supplies have been operating and running successfully on the Vulcan laser system since the system was inaugurated in 1975. But the equipment suppliers no longer manufacture or even support the electronics for the rod amplifiers. Therefore, it was necessary for the rod amplifier capacitors and control system to be replaced, as it posed a serious risk towards the future operation of Vulcan.

The Vulcan laser was shut down for three months from 12th August until 9th November to allow the replacement of the rod amplifier racks from inside the control room and laser areas with new capacitor banks.

New Quantel racks, located on north wall of Laser Area 1, to power the 45mm rod amplifiers and control processing and power of both rod chains.

The upgrade was successfully completed three days overdue and user experiments commenced on Monday 9th November 2009.

The experience gained from this project will aid the future success of the upcoming six-year 10 Petawatt upgrade project.



Old Quantel units inside the Vulcan Main Control Room.

Vulcan computer control system upgrade

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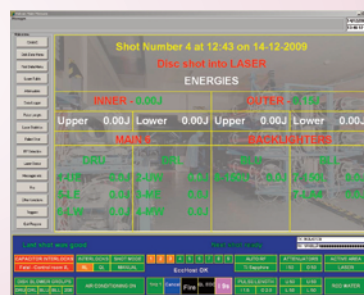
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The successful operation of the Vulcan high-power laser over the past 30 years has been heavily dependent on the use of computerized control systems. These originated with a GEC 4080 "mini" computer but in the mid 1980's were replaced with a pair of IBM 286 DOS-based PC's. The network continued to grow in size and complexity with (by the late 1990's) some peripheral PC's running Windows NT / 2000 but with the core fundamental laser control PC's remaining DOS.

A project to completely rewrite the computer control system, converting everything to windows XP compatible coding, turned out to be more complex than originally thought and became a four year project.



The bulk of the development for this took place offline with the final windows controls being tested, in parallel with the existing DOS controls, during the normal inter-experimental maintenance periods, a process which ensured minimal operational downtime.



Main control screen giving access to prime control functions and diagnostic data such as the shot firing sequence and waveform displays.

The Vulcan 10PW project – building design

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C.Biddle
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A.Gainer, J.Baker, R.Gibbs,
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(Nightingale Associates, Harwell, UK)

P.Ward, A.Hawkin
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M.Bacon, G.Rishworth, A.Pearson
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A key aspect of the Vulcan 10 PW project is the new building that will house the large amount of equipment required for the 10 PW laser facility. The building will extend the current building (R1) to maximise the available space within the limited surrounding land at RAL. In preparation for the start of phase two of the project a team consisting of STFC staff and external consultants has been working hard for the past six months to develop the

Vulcan 10PW building design. This resulted in a conceptual design report presentation and review.

Following this the 10PW management team have now agreed a conceptual design for the building that has been costed to a level adequate for the project to proceed to the building detail design, ready for anticipated tender action in the Autumn of 2010.



View of new 10 PW building.

Laser R&D

The DiPOLE project: towards high energy, high repetition rate diode pumped lasers



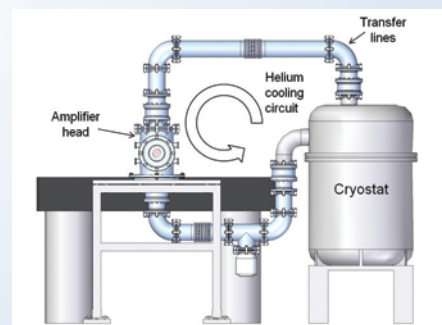
K. Ertel, S. Banerjee, P. Mason, J.P. Phillips,
P. Rice, S. Tomlinson, C. Sawyer, S.P. Blake,
C. Hernandez-Gomez, J.L. Collier
(CLF, STFC, Rutherford Appleton Laboratory, UK)

T. Davenne, M. Fitton, J. Hill, A. Lintern
(ETC, STFC, Rutherford Appleton Laboratory, UK)

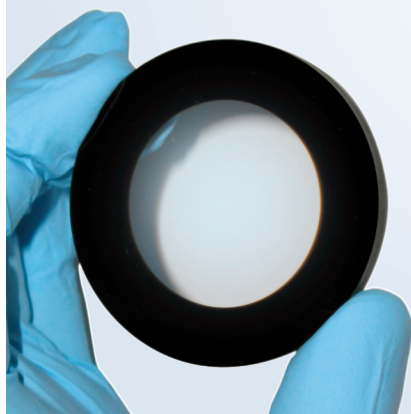
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DiPOLE stands for Diode Pumped Optical Laser for Experiments. It is a new project at the CLF to develop the foundations of novel high energy, high average power laser systems based on diode pumped solid state laser (DPSSL) technology. Compared to conventional systems, this approach promises dramatically increased repetition rates (and hence average powers) at significantly higher electrical-to-optical conversion efficiency. The article presents the conceptual design of a cryogenic Yb:YAG amplifier that can be scaled to kJ energy levels and beyond, owing to its geometry and unique cooling technique. Numerical modelling predicts superior performance of the amplifier at 175K compared to room temperature.

A distributed cooling technique using the slab-stack architecture aids in minimising amplified spontaneous emission (ASE) losses by enabling the use of a relatively thick gain media with low doping concentration. To test the concept in the laboratory, a lower-energy, multi-J prototype amplifier system is currently under development.



Schematic diagram of the cryocooler system under development at CLF.



Yb:YAG - Cr⁴⁺:YAG compound disk.

Timing and synchronisation system designs for the New Light Source



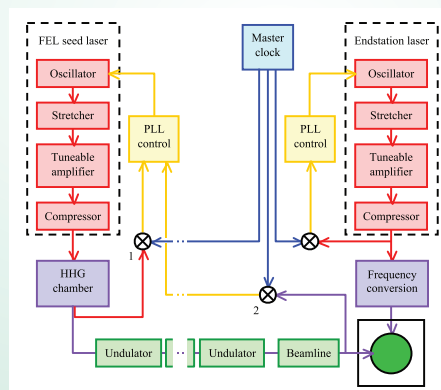
G.J. Hirst
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In recent years a new generation of “free electron laser” light sources has appeared, delivering beams which will revolutionise science just as conventional lasers have done since their invention 50 years ago.

The FELs, based on particle accelerators, are very large – many hundreds of metres long. But the light pulses they produce are very short. With a few tens of femtoseconds duration their physical length can be less than a hundredth of a millimetre. Much of the work that scientists want to do with them makes use of this very short duration, often combining beams from more than one ultrafast source in complex experimental geometries. Synchronising the pulses, which have travelled so far, so that they all arrive at exactly the right time on the target is a huge challenge. But a combination of ultra-low noise laser-based clocks, feedback-stabilised optical fibre transport and state of the art electronics promises to meet it!

A design for a system to synchronise light pulses from a free electron laser and a conventional laser at an experimental endstation.



Experimental setup in the Vulcan HaPPIE laboratory for multi-beam combination to achieve diffraction-limited pulses

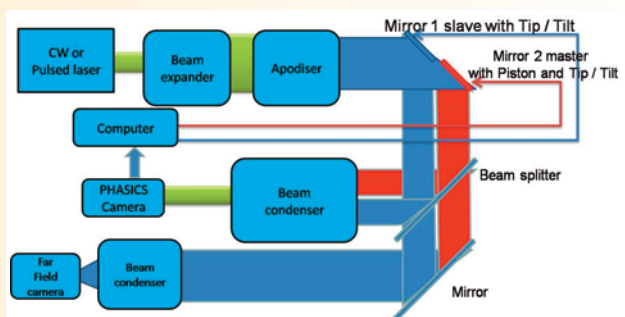


P.J. Phillips, C. Hernandez-Gomez, J.L. Collier
(CLF, STFC, Rutherford Appleton Laboratory, UK)

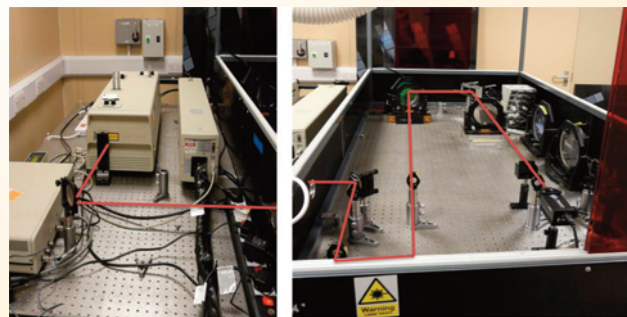
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With future and current designs for high power lasers requiring several sub-beams to be combined into a single monolithic one, technology needs to be developed to allow these beams to be spatially and temporally overlapped. A CLF laboratory has been set up - the Vulcan HaPPIE Laboratory (High Average and Peak Power lasers for Interaction Experiments) to research various techniques to spatially and temporally lock two or more sub-beams.

This article explains the various methods for one-aperture wavefront measurement and then goes on to explain how a phase difference (piston measurement) between two sub-apertures can be extracted using a proprietary technology. An experimental setup to test the concept is described and future developments are discussed.



Experimental layout.



Laboratory setup, beam path is shown by the use of the red line.

Target Fabrication

Installation of the target fabrication quality management system



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As target complexity is increasing and also with the introduction of high repetition rate laser systems such as Astra Gemini the demands on target fabrication to improve productivity and quality increase. It is essential to be able to trace, track and record target fabrication processes both during and after an experiment to interrogate data and to understand the underlying science that is achieved. To deliver this service and to improve and

streamline the processes from inception to delivery of a product the Target Fabrication Group has introduced a Quality Management System (QMS) into its experimental delivery procedures. This quality management process incorporates the full process of target fabrication from the initial conceptual design of a target, through the planning stage to manufacture and finally delivery to the experimental user group for their experimental shots.

Electroplating of gold and palladium for high power laser target fabrication



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Mass production of AFI cone geometries for fusion target studies



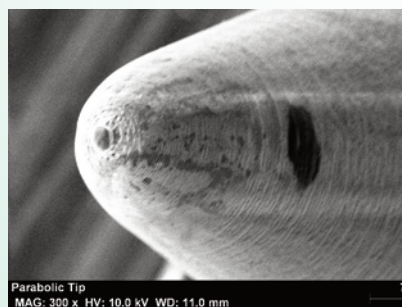
C. Spindloe, H.F. Lowe, M.K. Tolley
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P. Hiscock, M. Beardsley
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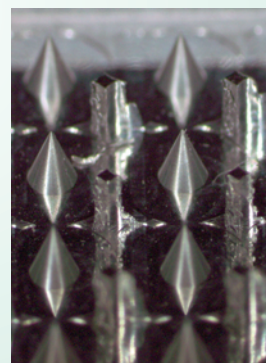
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One of the baseline targets for the HiPER project uses an innovation called AFI (Advanced Fast Ignition) in which a fuel sample is compressed using multiple laser beams around the end of a gold cone. These gold cones have been fabricated and assembled at RAL since 2003 using single 'one-off' production methods. Recently

mass production techniques have been developed that have dramatically reduced target costs, and therefore increase the availability of target components to the community. This initial work is summarised and more recent work to develop new target geometries and cones of different materials are discussed.



A SEM image of the parabolic shape of the tip.



An array machined in aluminium for copper plating

Instrumentation and Plasma Diagnostics

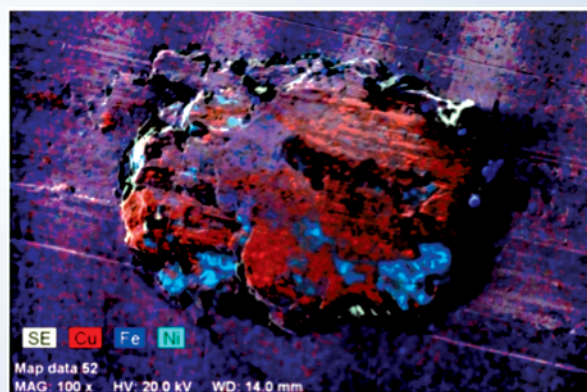
Debris analysis and mitigation for target motion systems



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(CLF, STFC, Rutherford Appleton Laboratory, UK)

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The use of motion stages within Target Area Interaction Chambers is key to the flexibility of experiments. Both targetry and diagnostics use such stages to allow in-vacuum positioning and adjustments in order to maximise the data output from shots. Failure of these systems has a high impact and usually requires the replacement of components in-situ. Over the last few years the 'sticking' encoders has dominated the failure modes for the standard motorised mounts. This report discusses the main cause of such sticking and one method for reducing the likelihood of reoccurrence.



EDX image of target debris coated onto the magnetic rod.

Calibration of image plate response to energetic carbon ions



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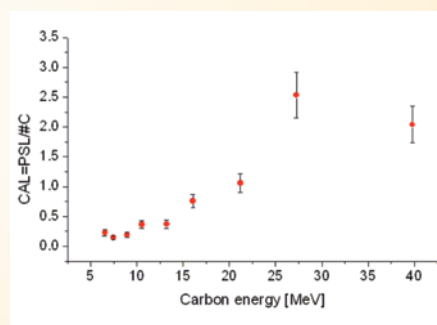
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Image Plate (IP) detectors are widely used for revealing x-rays, electrons and ions in laser-plasma experiments. While IP response to protons has been reported in earlier works, we have carried out for the first time measurements in which the IP response to Carbon ions has been measured by cross-calibration with CR39. In order to obtain the IP calibration for Carbon ions of different energies a Thomson Parabola is used to disperse the ion beam. A slotted CR39 placed on the IP is used as detector so that carbon ions having reasonably close energy can be recorded at same time around the edges of

the slots, both on the CR39 and IP. The PSL signal from the IP and the pits number on the CR39 were compared at a given ion energy.



Calibration curve that relates the PSL signal to the number of carbon ions as function of the ion energy.

Artemis Operational Statistics

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During the reporting year April 2009 - April 2010 a total of 5 experiments were delivered in the Artemis facility, which is under development, as shown in Table 1. At the user request the experiments were scheduled in recurring short, ~2 week slots. In total 21 experimental weeks were delivered to users excluding set-up weeks. 18 development weeks were allocated for the Artemis project. 3 weeks were allocated for the installation and commissioning of new equipment. 4 weeks were allocated to laser-services. Artemis facility overall availability was 142% when including the out of normal hours operations. Laser reliability was 88%.

Table 1. Artemis schedule 2009/10

Artemis	2009-2010	
	30-Mar-09	05-Apr-09
Marangos HHG with 2-colour laser Rapid Access	06-Apr-09	12-Apr-09
	13-Apr-09	19-Apr-09
	20-Apr-09	26-Apr-09
Artemis engineering XUV beamline	27-Apr-09	03-May-09
	04-May-09	10-May-09
	11-May-09	17-May-09
	18-May-09	24-May-09
	25-May-09	31-May-09
	01-Jun-09	07-Jun-09
	08-Jun-09	14-Jun-09
	15-Jun-09	21-Jun-09
	22-Jun-09	28-Jun-09
	29-Jun-09	05-Jul-09
Set-up week	06-Jul-09	12-Jul-09
Poletto - 90000 XUV beamline commissioning	13-Jul-09	19-Jul-09
	20-Jul-09	26-Jul-09
Mat Sci acceptance tests	27-Jul-09	02-Aug-09
Poletto - 90000 XUV beamline commissioning	03-Aug-09	09-Aug-09
	10-Aug-09	16-Aug-09
PI laser Service	17-Aug-09	23-Aug-09
Artemis engineering	24-Aug-09	30-Aug-09
	31-Aug-09	06-Sep-09
	07-Sep-09	13-Sep-09
Cavalleri – 90001 Material Science end station commissioning	14-Sep-09	20-Sep-09
	21-Sep-09	27-Sep-09

Artemis engineering	28-Sep-09	04-Oct-09
Red Dragon Laser Service	05-Oct-09	11-Oct-09
Artemis engineering	12-Oct-09	18-Oct-09
Cavalleri – 90001 Material Science end station commissioning	19-Oct-09	25-Oct-09
	26-Oct-09	01-Nov-09
Artemis engineering	02-Nov-09	08-Nov-09
	09-Nov-09	15-Nov-09
TOPAS laser installation: UV/Vis	16-Nov-09	22-Nov-09
Set-up week	23-Nov-09	29-Nov-09
Underwood 81003 Ultrafast dynamics of molecules	30-Nov-09	06-Dec-09
Red Dragon laser repair	07-Dec-09	13-Dec-09
	14-Dec-09	20-Dec-09
	21-Dec-09	27-Dec-09
Christmas and New Year	28-Dec-09	03-Jan-10
Red Dragon laser repair	04-Jan-10	10-Jan-10
	11-Jan-10	17-Jan-10
Red Dragon laser service	18-Jan-10	24-Jan-10
Artemis engineering	25-Jan-10	31-Jan-10
Cavalleri – 90001 Mat Science end station commissioning	01-Feb-10	07-Feb-10
Topas laser service	08-Feb-10	14-Feb-10
Petersen - 92000 EU Ultrafast dynamics in complex materials	15-Feb-10	21-Feb-10
Underwood 81003 Ultrafast dynamics of molecules	22-Feb-10	28-Feb-10
	01-Mar-10	07-Mar-10
	08-Mar-10	14-Mar-10
Petersen - 92000 EU Ultrafast dynamics in complex materials	15-Mar-10	21-Mar-10
	22-Mar-10	28-Mar-10
Hollow-fibre installation	29-Mar-10	04-Apr-10
Artemis engineering – vacuum system	05-Apr-10	11-Apr-10
Underwood 81003 Ultrafast dynamics of molecules	12-Apr-10	18-Apr-10

Table 2 and Figure 1 are an experiment by experiment break down of the Artemis facility performance for the reporting year and show a very good availability and reliability. The Artemis laser-beams were used for many long hours every day – while observing the standing orders for late-night working. 5 weeks around the end of 2009 were necessary for repairing the Red Dragon laser after one of the cryostats developed a vacuum leak: these weeks were also used for Artemis engineering in parallel to repairing the laser. This experimental time (Underwood experiment) was rescheduled in the New Year 2010 as shown in Table 1. The last week of this experiment was scheduled in April at the request of the user.

	Availability in normal hours (%)	Overall Availability (%)	Reliability (%)
Marangos	78	133	86
Poletto	81	142	88
Cavalleri	81	126	87
Underwood	75	141	85
Petersen	84	169	92

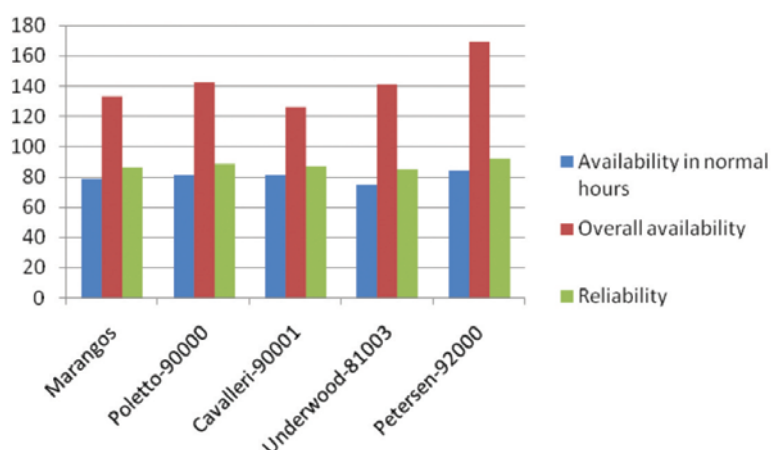


Figure 1. Artemis availability and reliability in 2009-10

Artemis proved very flexible in providing many combinations of fs pump-probe beamlines: Red Dragon, Topas (1300nm and UV), XUV beamlines (broadband and monochromatic), into end-stations: Materials Science end two visiting end-stations.

Astra Operational Statistics

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During the reporting year April 09 – April 10 a total of 3 complete experiments were delivered to the Astra-Gemini Target Area. In total 23 high power laser experimental weeks were delivered the Gemini target. The delivered schedule is presented below in figure 1.

The availability of the Gemini laser system (delivery to the Gemini Target Area) was 63% during normal working hours, rising to 104% with time made up from running out of normal working hours. The reliability of the Gemini laser was 73%. (represented in figure 3).

Gemini		
06-Apr-09	12-Apr-09	M Borghesi (LIBRA) 81025
13-Apr-09	19-Apr-09	
20-Apr-09	26-Apr-09	
27-Apr-09	03-May-09	
04-May-09	10-May-09	
11-May-09	17-May-09	
18-May-09	24-May-09	
25-May-09	31-May-09	
01-Jun-09	07-Jun-09	
08-Jun-09	14-Jun-09	
15-Jun-09	21-Jun-09	Contrast enhancement
22-Jun-09	28-Jun-09	
29-Jun-09	05-Jul-09	
06-Jul-09	12-Jul-09	
13-Jul-09	19-Jul-09	Macholite replacement and Amp 3 re-build
20-Jul-09	26-Jul-09	
27-Jul-09	02-Aug-09	Constrauction works
03-Aug-09	09-Aug-09	
10-Aug-09	16-Aug-09	Macholite replacement and Amp 3 re-build
17-Aug-09	23-Aug-09	
24-Aug-09	30-Aug-09	
31-Aug-09	06-Sep-09	
07-Sep-09	13-Sep-09	
14-Sep-09	20-Sep-09	Set up
21-Sep-09	27-Sep-09	
28-Sep-09	04-Oct-09	M Zepf 91009
05-Oct-09	11-Oct-09	
12-Oct-09	18-Oct-09	
19-Oct-09	25-Oct-09	
26-Oct-09	01-Nov-09	Jade 2 installation and contrast enhancement
02-Nov-09	08-Nov-09	
09-Nov-09	15-Nov-09	
16-Nov-09	22-Nov-09	
23-Nov-09	29-Nov-09	Coseners meeting
30-Nov-09	06-Dec-09	
07-Dec-09	13-Dec-09	Christmas
14-Dec-09	20-Dec-09	
21-Dec-09	27-Dec-09	Set up
28-Dec-09	03-Jan-10	
04-Jan-10	10-Jan-10	D Jarozynski 91032
11-Jan-10	17-Jan-10	
18-Jan-10	24-Jan-10	
25-Jan-10	31-Jan-10	
01-Feb-10	07-Feb-10	Contrast investigation and improvement
08-Feb-10	14-Feb-10	
15-Feb-10	21-Feb-10	
22-Feb-10	28-Feb-10	
01-Mar-10	07-Mar-10	Dual beam commisioning
08-Mar-10	14-Mar-10	
15-Mar-10	21-Mar-10	
22-Mar-10	28-Mar-10	
29-Mar-10	04-Apr-10	

Figure 1. Experiment by experiment breakdown of statistics

Following the Zepf experiment a number of changes were made to the Gemini operational model. Firstly the role of the designated Astra and Gemini operators were changed. The Astra operator now starts the Astra system up early, at 08:00 and brings up the Astra optical chain. The Gemini operator, as previously, brings up the Gemini systems, both operators then bring up full system shots. The Gemini operator now takes over sole responsibility for the system. The Gemini operator also maintains a presence at all times possible in the Gemini control room, such that there is a single local point of contact during the day. The presence of an operator at all time in control room means that system problems can be actively maintained and adjusted during the day rather than in a reactionary way to failures. There has also been an introduction of late operations cover 3 nights of the week, this allows the system to be maintained after 17:00 (a time identified when laser issues are more likely to occur) and into the early evening (as represented in figure 2 below). In order to improve communications the Astra operator for the following day meets briefly with the users at ~16:30 to discuss requirement for the next day.

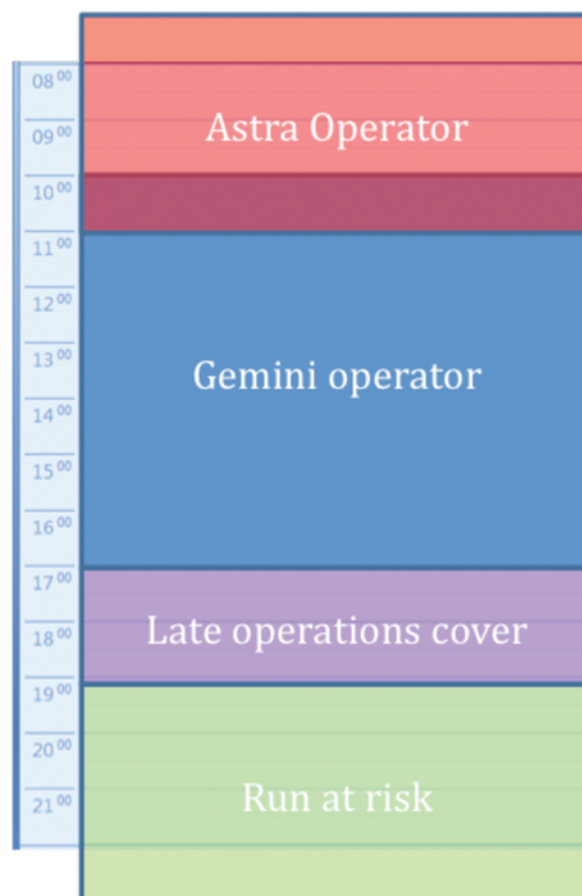


Figure 2. Revised Gemini operational model

During the reporting year a number of changes were made to the Astra systems in order to improve the system reliability. The major project during the year was to replace the macholite pump laser for the third Astra amplifier with 4 new Quanta Ray lasers. This project also involved a complete re-build of the amplifier. Previously the third amplifier straddled 2 optical tables, resulting in beam alignment stability issues. Additional optical tables were installed to accommodate the new pump lasers so the additional space created by the Macholite removal allowed the amplifier to be rebuilt on a single optical table. The beam expansion pipes for Gemini and ATA2 were lowered to bench height to improve access for optical alignment and to remove the need for periscopes, improving safety aspects of the amplifier.

The JADE pump laser for the KHz front was replaced with a more reliable JADE2. At the same time the Femtopower front end was reconfigured to give improved ASE contrast. A large number of aging optical components in the Astra chain were replaced with new optics with improved V-coatings. This removed a number of discrete picoseconds pre-pulses.

The Verdi pump laser for the Gemini oscillator was replaced with a new Verdi system; the replaced system had run for ~9000 hours without failure.

A number of periods of contrast characterization and improvement work were undertaken during the reporting year, the results of which are describe elsewhere in the annual report.

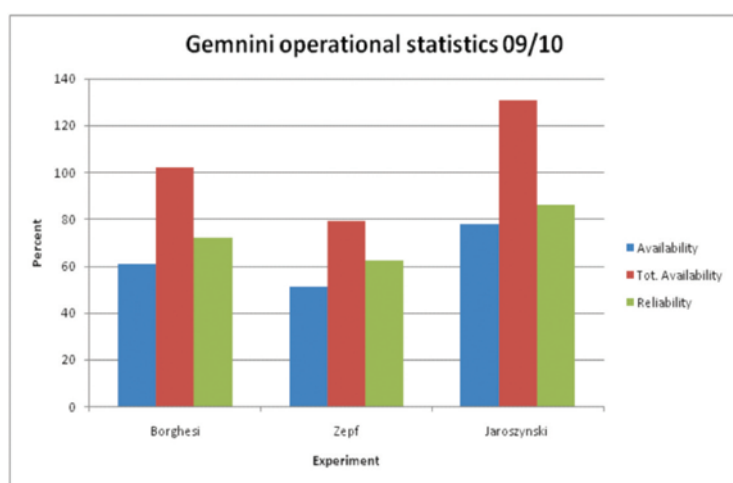


Figure 3. Experiment by experiment breakdown of statistics

Lasers for Science Facility Operational Statistics

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B.C. Coles, D.T. Clarke, I.P. Clark, A.W. Parker (CLF STFC, Rutherford Appleton Laboratory, UK)

RAL-based experiments

The reorganisation of the LSF has been completed with scheduling lines organised across the Functional Biological Imaging (FBI) and the Molecular Structural Dynamics (MSD). In response to requirements of the LSF to support cross Harwell Campus activity a line of resource has been put in place to reflect this and one can be confident that this section of the LSF will grow in the coming years as the LSF joins the Research Complex at Harwell. Both the MSD and FBI have made major new additions to their operational strengths. MSD turned on the BBSRC/STFC funded ULTRA laser system seeing its first User activity in July 2009, and following relocation of the single-molecule imaging capability from Daresbury to support the FBI biological applications of the LSF a new set of Users was welcomed in July. The year has also seen an enormous level of activity with the

division moving the suite of LSF laser systems into the Research Complex, the operation will be completed in September 2010. In the reporting period (April 2009 to March 2010), 21 different User groups performed a total of 23 experiments in the LSF laboratories at RAL. A total of 3110 hours laser time was delivered to the UK User community and European Users throughout the year, with only 46 hours downtime. The majority subject group scheduled was biological related science, see figure 1. A full breakdown by subject number of weeks applications versus weeks scheduled is shown in figure 2 showing a 2 - 3 times level of oversubscription. The RAL-Based schedule is shown in table 1. The average User satisfaction marks obtained from the scheduled Users are shown in figure 3. There were a total of 20 formal reviewed publications produced from the years efforts, with the LSF programme supporting 4 students working towards a PhD in the reporting year.

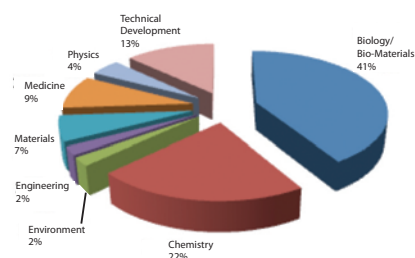


Figure 1. RAL-based bids by subject group

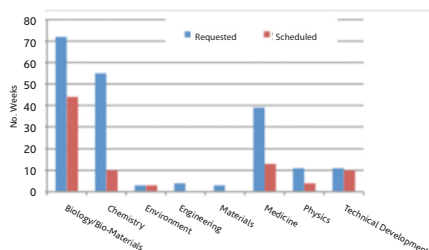


Figure 2. RAL-based experiments by subject

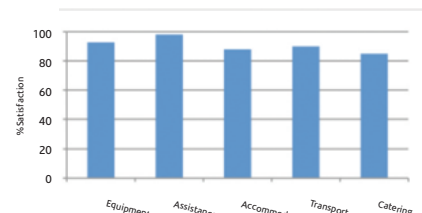


Figure 3. RAL-based average User satisfaction scores

Loan Pool

In March 2009 the new EPSRC Loan Pool grant commenced, this is for £2.6m over 5 years and will see the replacement of many of the laser systems within the facility. This year saw the introduction of a Steering Committee to the Loan Pool which will meet annually to provide advice on direction and purchases for the facility. Following the first meeting a number of purchases are in progress many of which will enable the facility to target a broader community. The delivery of the first system a quasi-cw supercontinuum source is expected in summer 2010. Further details on the developments can be found in the article

“Developments within the EPSRC Laser Loan Pool”.

The Loan Pool delivered 324 weeks of laser time in the reporting period with a ratio of weeks applied for to weeks scheduled of 1.33:1. Downtime was 32 weeks which is unusually high; the vast majority of this was for a fault with the UFL2 system which required two factory repairs before the fault was rectified, losing an entire loan period. The year's activity saw 4 new research groups use the Loan Pool. The chemistry community was once again the biggest User with 33% of allocated time. The breakdown is shown in figure 4. The Loan Pool schedule is shown in table 2. There were a total of 8 publications.

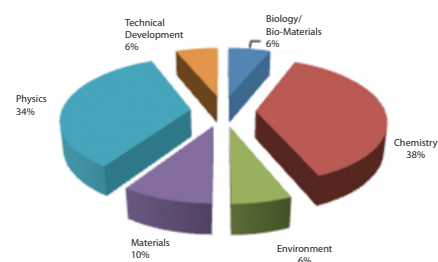


Figure 4. Loan Pool bids by subject group

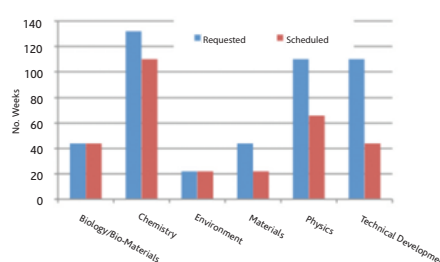


Figure 5. Loan Pool experiments by subject

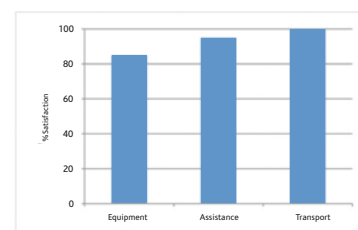


Figure 6. Loan Pool average User satisfaction scores

Table 1. Lasers for Science Facility RAL-based Schedule 2009/10

	Date	Functional Biosystems Imaging	Molecular Structural Dynamics	Cross Department Research
2009	6 Apr	RELOCATION OF ASMID TO RAL	ULTRA SETUP AND DEVELOPMENT	
	13 Apr			
	20 Apr			
	27 Apr			
	4 May			
	11 May			
	18 May			
	25 May			
	1 Jun			
	8 Jun			
	15 Jun			
	22 Jun			
	29 Jun			
	06 Jul		M SEARLE (University of Nottingham) DNA Hairpin Folding by 2DIR 91033 Proof of concept proposal for ULTRA	
	13 Jul		N SCRUTTON (University of Manchester) Dynamics of Light Activated Enzyme POR 91033 Proof of concept proposal for ULTRA	Currel/Botchway DLS-MT106
	20 Jul	M MARTIN-FERNANDEZ (STFC) 91,011 Supra-molecular rules in signalling networks		
	27 Jul		M SEARLE (University of Nottingham) DNA Hairpin Folding by 2DIR 91033 Proof of concept proposal for ULTRA	
	03 Aug		M GEORGE (University of Nottingham) Infrared Coherent Control 91033 Proof of concept proposal for ULTRA	
	10 Aug	A W PARKER (STFC) 91,032 Studies to unravel the specific protein signalling pathways	SETUP AND MAINTENANCE	A D WARD (STFC) 91,038 X-ray diffraction studies from microcrystals
	17 Aug			
	24 Aug	M MARTIN-FERNANDEZ (STFC) 91,011	C JOHNSON (MRC-LMB Cambridge) Protein Folding of BBL 91033 Proof of concept proposal for ULTRA	
	31 Aug	A W PARKER (STFC) 91,032	M GEORGE (University of Nottingham) Non-statistical Dynamics 91033 Proof of concept proposal for ULTRA	A D WARD (STFC) 91,034 Target delivery using optical levitation
	07 Sept		A J ORR-EWING (University of Bristol) 91,004 Dynamics of chemical and photochemical reactions in solution	
	14 Sept			
	21 Sept	M MARTIN-FERNANDEZ (STFC) 91,011	A W PARKER (STFC) 91,031 Investigating the role of secondary and tertiary structure on DNA photostability	

	28 Sept	C HAWES (Oxford Brookes University) 91,014 The plant secretoryome	A W PARKER (STFC) 91,031 Investigating the role of secondary and tertiary structure on DNA photostability	
	05 Oct			
	12 Oct			M MARTIN-FERNANDEZ (STFC) 91,011
	19 Oct	A JESHTADI (Oxford Brookes University) 91,025 Autographa californica multinucleopolyhedrovirus Capsid protein	MAINTENANCE	
	26 Oct	A W PARKER (STFC) 91,032	M TOWRIE (STFC) 91,022 A novel hybrid Raman-IR technique	
	02 Nov			J C HILLER (DLS) 91,015 A ground-breaking dual beam laser trap for X-ray experimental facilities
	09 Nov			J R DILWORTH (Oxford University) 91,009 Evaluating the intracellular stability of complexes of metallic radionuclides
	16 Nov	M MARTIN-FERNANDEZ (STFC) 91,011		
	23 Nov		MAINTENANCE	A D WARD (STFC) 91,034
	30 Nov		P O'NEILL (Oxford University) 91,035 Dynamics of genomic DNA damage repair proteins	
	07 Dec	MAINTENANCE		
	14 Dec	M MARTIN-FERNANDEZ (STFC) 91,011!		
	21 Dec	CHRISTMAS AND NEW YEAR		
	28 Dec			
2010	4 Jan	MAINTENANCE	MAINTENANCE	A D WARD (STFC) 91,034
	11 Jan		P PORTIUS (University of Sheffield) 91,036 Real-time structural dynamics of molecular systems for energy generation and storage	
	18 Jan	J R DILWORTH (Oxford University) 91,009		
	25 Jan	A JESHTADI (Oxford Brookes University) 91,025	N T HUNT (University of Strathclyde) 91,003 Dynamics and reactivity of the FeFe[Hydrogenase] enzyme system	Currel/Botchway DLS-EE106
	01 Feb	J R DILWORTH (Oxford University) 91,009		
	08 Feb	C HAWES (Oxford Brookes University) 91,014		
	15 Feb	A JESHTADI (Oxford Brookes University) 91,025		
	22 Feb	M MARTIN-FERNANDEZ (STFC) 91,011	J M KELLY (University of Dublin) 92,005	
	01 Mar	P O'NEILL (Oxford University) 91,035	M T PRYCE (Dublin City University) 92,004	
	08 Mar	MAINTENANCE		
	15 Mar	S J QUINN (University College Dublin) 101,002 I CASIMIRO (Universidad de Extremadura) 101,000	P PORTIUS (University of Sheffield) 91,036	
	22 Mar	P O'NEILL (Oxford University) 91,035		
	29 Mar	S J QUINN (University College Dublin) 101,002	MAINTENANCE	

Table 2. Lasers for Science Facility Loan Pool Schedule 2009/10

	Date	NSL1	NSL2	NSL3	NSL4	NSL5	UFL1	UFL2	CWL1
2009	Apr 6	HIPPLER (Sheffield) High-resolution stimulated Raman spectroscopy with photo-acoustic detection (PARS) 82,008	SIMONS (Oxford) Carbohydrate molecular recognition: Probing CH- π interactions 82,007	RUDDOCK (Strathclyde) Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing 82,005	SNOEK (Oxford) Studying the influence of metal cation binding on zinc finger moldel peptide folding behaviour 82,004		JONES (Edinburgh) 2-photon excitation microscopy and confocal FLIM 81,011	BENNINGTON (STFC) Using tip enhanced femtosecond lasers to create graphite nano-structures on diamond 82,010	DUTTON (STFC) Surface Raman spectroscopy of photo-catalytic hydrogen production processes 82,001
	Apr 13								
	Apr 20								
	Apr 27								
	May 4								
	May 11								
	May 18								
	May 25								
	Jun 01								
	Jun 08								
	Jun 15								
	Jun 22								
	Jun 29								
	Jul 06								
	Jul 13								
	Jul 20								
	Jul 27								
	Aug 03								
	Aug 10								
	Aug 17								
	Aug 24								
	Aug 31								
	Sep 07								
	Sep 14								
	Sep 21								
	Sep 28								
	Oct 05								
	Oct 12								
Oct 19									
Oct 26									
Nov 02									
Nov 09									
Nov 16									
Nov 30									
Dec 07									
Dec 14									
Dec 21									
Dec 28									
Dec 28									
2010	Jan 04	RITCHIE (Oxford) Measurement of fragment polarization from photolysis of vibrationally state prepared molecules 92,000			Repair		CLAEYSSSENS (Sheffield) 2-photon 3D structuring of photo-curable biopolymers 91,027	WU (York) Effect of laser pulse width/energy on optically-induced magnetisation reversal 91,007	VOLK (Liverpool) Polymer coil-globule transition dynamics on the ms-time scale 91,024
	Jan 11								
	Jan 18								
	Jan 25								
	Feb 01								
	Feb 08								
	Feb 15								
	Feb 22								
	Mar 01								
	Mar 08								
	Mar 15								
	Mar 22								
	Mar 29								

Target Fabrication Operational Statistics

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RAL Experiments

A total of one Astra Gemini and eight Vulcan experiments were supported by Target Fabrication in the reporting period April 2009 to April 2010. All these experiments were 'solid target' experiments. Target Fabrication provided a total of 49 weeks of experimental support for Vulcan and 4 weeks for Astra Gemini on solid target experiments. The report does not include the extensive amount of filter and pinhole support provided from Target Fabrication for some jet experiments.

1) Target Numbers

For the reporting year, the total target numbers produced are shown in Table 1. The table is broken down into separate experiments and gives data on total target numbers produced and the subset consisting of high specification complex 3D targets that have been produced. High specification 3D targets are defined as targets that have taken significant highly skilled microassembly or micromachining to be produced over and above standard target manufacture. The total number of targets for use at RAL produced by the group in 2009-2010 was 1,424 compared to 1,023 in 2008-2009¹. During 2009-2010, the number of high specification targets produced was 379 accounting for 27% of the total targets made which is consistent with the figure of 29% for 2008-2009¹.

Experiment	Targets Produced	High Specification Targets
May 2009 TAP	143	2
May 2009 TAW	116	48
July 2009 TAP	101	53
July 2009 TAW	197	82
Oct 2009 GTA	97	0
Nov 2009 TAP	60	43
Nov 2009 TAW	153	56
Jan 2010 TAW	274	95
Mar 2010 TAW	283	0
TOTAL	1424	379

Table 1: Target production summary for 2009-2010.

2) Target Types

The high specification targets can be separated into 6 main types as shown in Table 2 and Figure 1. Most notably, 53 multilayered microsquare targets were delivered to the July 2009 TAP experiment and 95 thin wire targets were delivered to the January 2010 TAW experiment requiring extensive highly skilled microassembly.

Target Type	Targets Produced
Multilayered Microsquares (100um x 100um)	53
XRTS	41
Multilayered Opacity Targets	28
Thin Wires	95
Ultra Thin DLC	35
Other	0

Table 2: High Specification Target Delivery Summary.

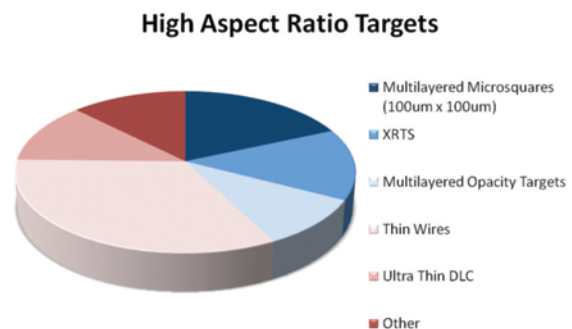


Figure 1: High Specification Target Delivery Summary.

3) Experimental Response

It is seen as a significant strength of Target Fabrication to be rapidly responsive to experimental results and conditions by working collaboratively with experimental groups. The Target Fabrication group responds to experimental changes during a run and often implements a number of modifications or redesigns to the original requests. The number of modifications and variations usually fluctuates widely across a year and is dependent on the type of experiment and also on experimental conditions such as diagnostic and laser performance. On average, during experiments in the reporting period, 42% of the targets that were shot were modified or redesigned from the planned target specifications. This is higher than the figures for the previous 2 years, which were 30% in 2008-2009¹ and 32% in 2007-2008². Figure 2 shows the proportion of targets that were redesigned or modified during the experiments supported in 2009-2010.

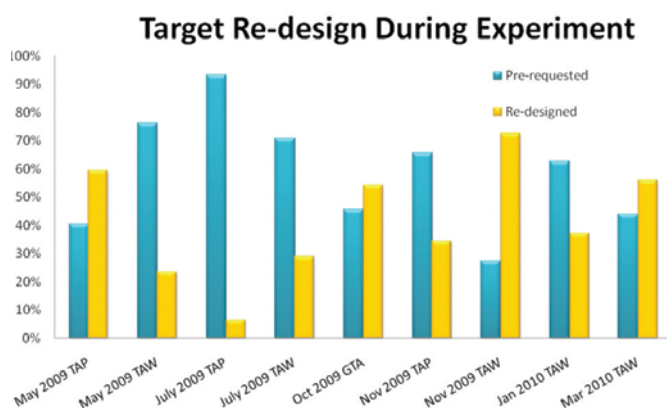


Figure 2: The percentage of pre-requested targets compared to redesigned targets fabricated throughout the year for each experiment.

The redesign of targets during experiments means that there are often a number of targets that have been fabricated but that are not shot by the end of experimental campaigns. As shown in Figure 3, throughout the year an average of 39% of targets that were fabricated were either returned un-shot to Target Fabrication or were unused having been made in preparation for the experiment but not required due to changes. This is slightly higher than the figures of 35% for 2008-2009¹ 36% for 2007-2008².

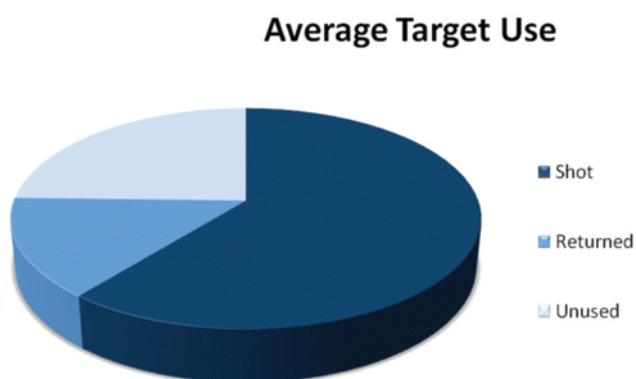


Figure 3: The average proportion of targets shot, returned and unused during solid target experiments on Vulcan in 2009-2010.

External Contracts

Scitech Precision Ltd, a spinout company from Target Fabrication, also supplied microtargets, specialist coatings and consultancy to a number of external contracts. A total of ten experiments were supported at external facilities including LULI, PALS, GEKKO and GSI.

Summary

Target Fabrication has supported nine internal and ten external experimental groups in the last year as well as providing an increasing amount of characterisation services and acting as a knowledge base for Target Fabrication activities throughout Europe. There was an increase in the number of targets delivered to experiments on Vulcan compared to 2008-2009 even though the number of experiments scheduled was the same.

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Vulcan Operational Statistics

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Introduction

Vulcan has completed an active experimental year, with 48 full experimental weeks allocated to target areas TAW and TAP between April 2009 and March 2010. This figure is down on previous years for two reasons :

- 1) The Vulcan laser was shut down for three months from August to November 2009 for the rod amplifier upgrade and recommissioning¹.
- 2) Single area operations were adopted in January 2010 to enable front end tests to be undertaken.

Table 1 below shows the operational schedule for the year, and illustrates the shot rate statistics for each experiment. Numbers in parentheses indicate the total number of full energy laser shots delivered to target, followed by the number of these that failed.

PERIOD	TAW	TAP
4 May – 14 Jun	G Tallents EUV probing of solar opacity (41, 5) (87.8%)	M Borghesi Plasma jets driven by petawatt pulses (48, 7) (85.4%)
29 Jun – 9 Aug	J Fuchs Influence of overlapping high-intensity laser beams on electron and ion generation and transport (83, 10) (88.0%)	M Zepf Measurement of enhanced coupling in counter-propagating fast electron beams interaction in high-intensity laser-plasma experiments (42, 12) (71.4%)
10 Aug – 8 Nov	Rod amplifier upgrade	
9 Nov – 20 Dec	D Riley Short range screening in warm dense matter (51, 5) (90.2%)	Z Najmudin Ion acceleration from ultra-thin targets and near critical density targets (43, 8) (81.4%)
11 Jan – 21 Feb	Hoarty (AWE) A proposal to use laser produced proton beams to isochorically heat materials for warm dense matter studies (75, 10) (86.7%)	
8 Mar – 18 Apr	M Borghesi Laser-driven ion-acoustic solitons in tenuous plasmas (62, 8) (87.1%)	

Table 1. Experimental schedule for the period April 2009 – March 2010

(Shots fired, failed shots)
(Reliability)

The total number of full disc amplifier shots that have been fired to target this year is 445. This is a decrease compared with recent years (Table 2) for the reasons given above. 65 shots failed to meet user requirements. The overall shot success rate to target for the year is 85%, compared to 91%, 90%, 85%, 86% in the previous four years. Figure 1 shows the reliability of the Vulcan laser to all target areas over the past five years.

The shot reliability to TAW has remained roughly constant compared with 2008-2009 at 88%. This is encouraging considering that all of the experiments in TAW in this reporting year were carried out after the upgrade. The shot rate in Vulcan has dropped this year due to the shutdown for the rod amplifier upgrade. Again following this upgrade the overall reliability of Vulcan has remained unaffected. The shot reliability to TAP is around 80% and efforts in the coming year will be focused on increasing the number of good shots to TAP.

Analysis of the failure modes reveals that, as in recent years, the two overriding causes of failed shots are alignment and front end related issues. It is difficult to distinguish these two causes and in the coming year we will be commissioning further diagnostics on the rod amplifier chain (energy monitors and beam profiling after each amplifier). Issues with the front end system over the past year have included timing issues with the shaped long pulse oscillator, the spectrum of the silicate / phosphate chain and the opcpa pump laser.

	No of shots	Failed shots	Reliability
05 - 06	672	93	86%
06 - 07	1043	149	85%
07 - 08	977	98	90%
08 - 09	646	61	91%
09 - 10	445	65	85%

Table 2. Shot totals and proportion of failed shots for the past five years

There is a requirement which was originally instigated for the EPSRC FAA that the laser system be available, during the four week periods of experimental data collection, from 09:00 to 17:00 hours, Monday to Thursday, and from 09:00 to 16:00 hours on Fridays (a total of 156 hours over the four week experimental period). The laser has not always met the startup target of 9:00 am but it has been common practice to operate the laser well beyond the standard contracted finish time on several days during the week. In addition, the introduction of early start times on some experiments continues to lead to improvements in availability.

On average, Vulcan has been available for each experiment to target areas for 76.4% of the time during contracted hours and 105.6% overall. These figures compare with 78.8% and 108.2% in 2008-2009 to all target areas. The time that the laser is unavailable to users is primarily the time taken for beam alignment at the start of the day. Over the past twelve months, each experiment has also experienced an average of 5.7 hours during the standard working week when the laser has been unavailable, or just over one hour per day.

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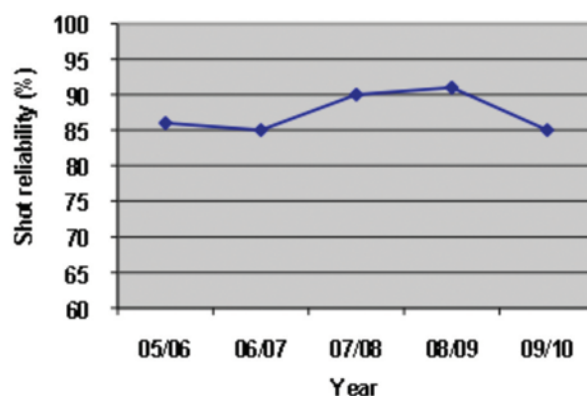


Figure 1. All areas shot reliability for each year 2005-6 to 2009-10

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Thesis

HIGH POWER LASER FACILITY – ASTRA & VULCAN

B. Ramakrishna

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P. Wilson

Studies of laser generated relativistic electron dynamics using proton probing

PhD Thesis, Queens University, Belfast 2009

D. Carroll

Laser-driven ion acceleration : source optimisation and optical control

PhD Thesis, University of Strathclyde, 2009

S. Nagel

Studies of Electron Acceleration Mechanisms in Relativistic Laser-Plasma Interactions

PhD Thesis, Imperial College, London (2009)

C. Bellei

Measurements of Optical Radiation from High-Intensity Laser-Plasma Interactions

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C. Kamperidis

Investigation of Electron Laser Wakefield Acceleration in Novel Plasma Structures

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Rashida Jafer

Laser Based proton generation and proton radiography of laser driven cylindrical implosion

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K. Quinn

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LASERS FOR SCIENCE FACILITY

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Investigation of cellular DNA damage induced by multiphoton laser light absorption of near infrared femtosecond laser pulses

PhD Thesis, Reading University (2009)

Panel membership and CLF structure

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Dr D. T. Clarke (Functional Bio-systems Imaging)
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Vulcan, Astra TA2 & Gemini Facility Access Panel 2009/10

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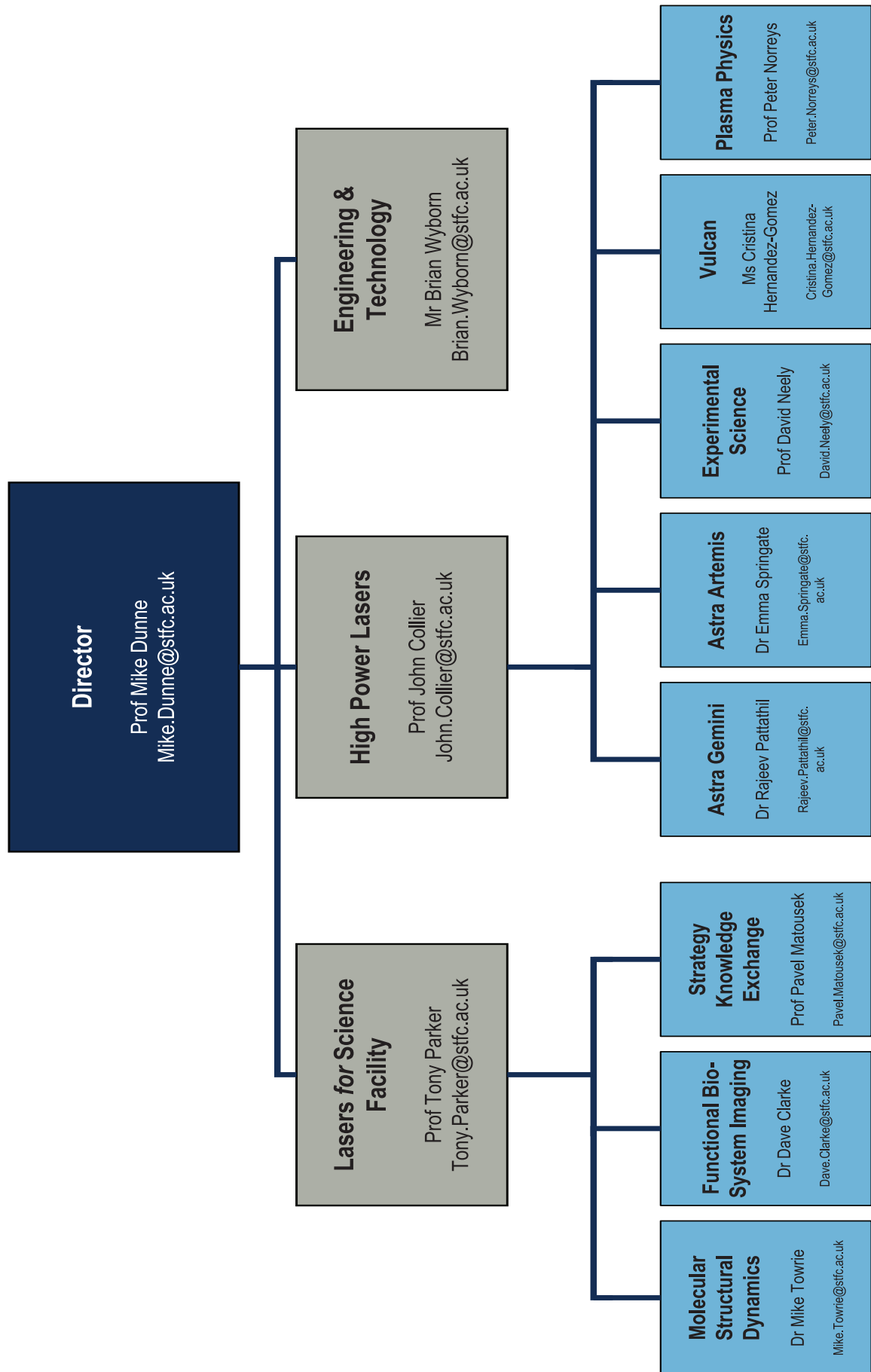
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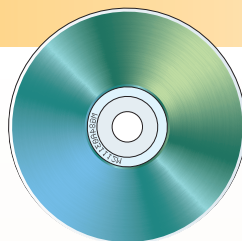
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CENTRAL LASER FACILITY STRUCTURE 2009/10





CD Rom Index

High Power Laser Science

Plasma Physics

- 1 Plasma jets and narrow bandwidth ion spectra from thin foils irradiated at high laser intensity
- 2 Influence of overlapping high-intensity laser beams on electron and ion generation and transport
- 3 Characterization of a line focus back-lighter
- 4 Creation of persistent, straight, 2mm long laser-driven channels in underdense plasmas for fast ignition applications
- 5 Observation of post-soliton expansion following laser propagation through an underdense plasma
- 6 Perspectives on radiative blast waves in laser-heated clustered gases

Femtosecond Pulse Physics

- 7 Radiation pressure effects on ion acceleration on the Gemini laser
- 8 Ion acceleration from foil targets in the ultraintense ultrahigh contrast regime.
- 9 Characterisation of debris emission from thick targets on Astra-Gemini
- 10 High rep rate Thomson parabola-MCP assembly for multi-MeV ion spectroscopy
- 11 Proton/ion energy scaling and laser conversion efficiency using 50fs, 10^{20} - 10^{21} W/cm² Astra-Gemini pulses
- 12 Investigation of contrast of Astra/Gemini

Theory & Computation

- 13 Electron transport during shock ignition
- 14 New perspectives on the electrodynamics of intense laser-plasmas
- 15 Cavitation and Shock Wave Formation in Dense Plasmas by Relativistic electron beams
- 16 Alpha-particle stopping power for dense hydrogen plasmas
- 17 An object-oriented 3D view-factor code for hohlraum modelling
- 18 Calculation of Siegert states in electric field: from atoms to molecules
- 19 Finite size effects in high-intensity QED
- 20 Multi-electron dynamics on the femtosecond time-scale

- 21 Hydrodynamics of the Yukawa one component Plasma

- 22 The accuracy of rear-surface measurements of the angular-divergence of a laser-generated fast-electron beam

- 23 High absorption of ultra-intense laser pulses in near critical plasmas

- 24 Modelling Spitzer transport in laser produced plasmas by direct Vlasov methods with a BGK collision operator

- 25 Effect of channel profile evolution on laser-driven electron acceleration in plasma channels

- 26 A Vlasov-Fokker-Planck code for shock ignition

- 27 Multi-component effects on the x-ray scattering signal from warm dense matter

Ultrafast and XUV Science

- 28 Quasi-classical model of non-destructive wavepacket manipulation by intense few-cycle nonresonant laser pulses

- 29 Monochromatic XUV-photon + strong-field NIR cross-correlation by atomic excitation and ionization

- 30 Multi-pulse scheme for controlling electron localisation upon molecular dissociation

- 31 Intensity-resolved ionization processes in few-cycle strong-field laser pulses

- 32 Ultrafast dynamics of electronic structure in complex materials

- 33 Ultrafast time-resolved photoelectron imaging of excited state molecular dynamics: S_2 - S_1 internal conversion in the DABCO molecule

Lasers for Science Facility Programme

Biology

- 34 Probing metal complex dissociation in cells by fluorescence microscopy

- 35 Biocompatible microstructured materials for tissue engineering

- 36 The plant secretoryome: protein-protein interactions in the higher plant secretory pathway

- 37 Supra-molecular rules in signalling networks: A single molecule comparative study in cells and tissues

- 38 The role of BER proteins in the repair of DNA damage induced following NIR multiphoton laser microbeam irradiation



Chemistry

- 39** Time resolved infrared analysis of AppABLUF using isotopic labeling at mutant studies
- 40** High-resolution stimulated Raman spectroscopy with photoacoustic detection (PARS) of formic acid dimer
- 41** Transient 2D-IR spectroscopy of [FeFe]hydrogenase enzyme model compounds
- 42** Ultrafast TRIR studies on tetraazidosilicon complexes
- 43** Photoinitiated CO-release molecules
- 44** Excited-state dynamics of adenine thymine dinucleotides: influence of stacking
- 45** Antigens laid bare: the intrinsic conformation of Lewis^x
- 46** Carbohydrate molecular recognition: probing CH- interactions
- 47** Polymer coil-globule transition dynamics on the nanosecond to second time scale

Physics

- 48** Using tip enhanced femtosecond lasers to create graphite nanostructures on diamond
- 49** Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing
- 50** Distributed sensing by time-correlated two-photon excited fluorescence in rare earth doped optical fibres

Laser Science and Development

Artemis

- 51** Monochromatised XUV beamline for ultrafast time-resolved ARPES
- 52** Spin and Angle resolved photoemission with fs laser source: calibration of spin detector
- 53** Few-cycle carrier-envelope-phase controlled laser pulses for time resolved science at the Artemis facility

Astra

- 54** Vibration in Gemini and Engineering modifications (interim report)
- 55** Temporal and spatial overlap monitoring of the dual-beam layout in Astra-Gemini TA3
- 56** Upgrade of Astra Amplifier 3 and the Astra interlock system
- 57** Improved post-experiment data analysis at Astra Gemini
- 58** A new phase (and amplitude) for Astra Gemini: using a spatial light modulator
- 59** Ultrafast gated imaging of laser produced plasmas using the optical Kerr effect

Lasers for Science Facility

- 60** Two-photon fluorescence correlation spectroscopy: FCS
- 61** The ULTRA time-resolved IR, 2D-IR and T-2D-IR station
- 62** Developments within the EPSRC Laser Loan Pool
- 63** The OCTOPUS Imaging Cluster: A New Imaging Facility in the Research Complex at Harwell
- 64** New data analysis software for time-resolved spectroscopy experiments in the ULTRA laser facility
- 65** Active synchronisation of dual amplifier outputs for time-resolved spectroscopy
- 66** Optical trapping of laser targets under vacuum for ion-beam production in the LIBRA programme

Vulcan

- 67** Close-in contrast measurements of the new ps OPCPA front end
- 68** Vulcan rod amplifier upgrade
- 69** Vulcan computer control system upgrade
- 70** The Vulcan 10PW project – building design

Laser R&D

- 71** The DiPOLE project: towards high energy, high repetition rate diode pumped lasers
- 72** Timing and Synchronisation System Designs for the New Light Source
- 73** Experimental setup in the Vulcan HaPPIE Laboratory for Multi-beam Combination to achieve Diffraction limit pulses

Target Fabrication

- 74** Installation of the target fabrication quality management system
- 75** Electroplating of Gold and Palladium for High Power Laser Target Fabrication
- 76** Mass production of AFI cone geometries for fusion target studies

Instrumentation and Plasma Diagnostics

- 77** Debris analysis and mitigation for target motion systems
- 78** Calibration of image plate response to energetic carbon ions

Author Index

A

Ahlswede, H.C.	37
Alexander, C.	30
Alexander, J.D.	22
Allen, P.	42
Arber, T.D.	18
Arrowsmith, R.L.	24
Audebert, P.	7

B

Bacher, A.	27
Bacon, M.	42
Baeva, T.	10
Banerjee, S.	43
Baker, J.	42
Barber, E.	45
Beardsley, M.	45
Bell, A.R.	13, 19
Belshaw, L.	22
Bennington, S.M.	31
Bentley, C.	30
Berger, H.	23
Bewley, R.I.	31,
Biddle, C.	42
Blake, S.P.	35, 42, 43
Bonora, S.	33
Booth, N.	8, 35
Borghesi, M.	7, 8, 9, 10, 11, 12, 46
Botchway, S.W.	24, 25, 26, 38
Boyle, A.G.	41
Brenner, C.M.	10, 11, 12
Brust, R.	27
Bryan, W.A.	21, 22, 33, 34
Burgos, P.	38
Burton, D.A.	13
Bush, I.	14
Buffechoux, S.	7

C

Cacho, C.M.	23, 33, 34
Carroll, D.C.	10, 11, 12
Calvert, C.R.	21, 22
Cameron, F.H.	10
Cavalieri, A.	23, 33
Cavalleri, A.	33
Cerchez, M.	7, 46
Chekhlov, O.V.	12
Claeysens, F.	24
Clark, I.	27
Clark, I.P.	28, 38, 39, 51
Clark, R.J.	7
Clarke, D.T.	25, 38, 51
Clarke, R.J.	11, 42
Clarke, R.	46
Clark, E.L.	8, 9
Cocinero, E.J.	29, 30
Coles, B.C.	51
Collier, J.L.	42, 43, 44
Cooper, S.L.	26
Costello, B.	36, 41
Cox, A.	46

D

Daligault, J.	17
Dalzell, C.J.	31, 32
Davis, B.G.	30
Davies, J.R.	8, 9
Davenne, T.	43
Dean, N.	23, 33
Dhesi, S.	23, 33
Dilworth, J.R.	24
Divall, E.J.	37
Doorley, G.W.	29
Doria, D.	7, 10, 11, 12, 46
Dover, N.P.	10, 11, 12
Doyle, H.W.	9

E

East, I.	45
Edie, D.J.	14
Ellison, L.	7
Ernst, B.	29
Ertel, K.	12, 43
Evans, R.G.	17

F

Faulkner, S.	24
Fisher, M. J.	15
Fitton, M.	43
Fonseca, R.A.	19
Foster, P.S.	10, 11, 12, 35
Frackiewicz, A.	41
Frassetto, F.	21
Frassetto, F.	33
Froud, C. A.	21, 22, 33
Fuchs, J.	7

G

Galimberti, M.	41
Gallegos, P.L.	10, 11, 12, 37
Gainer, A.	42
Gartside, L.M.R.	8
Gericke, D.O.	14, 20
Gibbs, R.	42
Gil, J.M.	9
Gill, A.A.	24
Green, J.S.	10, 11, 12
Greenwood, J.B.	21, 22
Greetham, G.M.	27, 28, 39
Gregori, G.	17, 20
Gumbrell, E.T.	9

- H**
- Haigney, A. 27
- Hamonou, L. 15
- Han, T.P.J. 31, 32
- Hancock, S. 42
- Harvey, E.C. 28
- Hassan, S. 8, 9
- Hawes, C. 25
- Hawkes, S. 12, 36, 49
- Hawkin, A. 42
- Heathcote, R. 7, 8, 41
- Heinzl, T. 16
- Henstridge, J.M. 42
- Hernandez-Gomez, C. 41, 42, 43, 44
- Hippler, M. 27
- Hirsh, M. 25, 38
- Hirst, G.J. 43
- Hiscock, P. 45
- Hill J. 43
- Holligan, P. 42
- Hohenberger, M. 9
- Holland, D.M.P. 23
- Hollis, D.B. 32
- Hook, S.G. 21, 22, 33, 46
- Hooker, C.J. 12, 36, 37
- Huang, H. 35
- Hunt, N.T. 28
- Hutchinson, S. 16
- I**
- Ilderton, A. 16
- J**
- Jackmann, R.B. 31
- Jiang, J. 8, 9
- John, C. 36
- Jones, M.W. 24
- K**
- Kageiwa, N. 8, 9
- Kaiser, S. 23
- Kakolee, K. 7, 46
- Kamperidis, C. 19
- Kar, S. 7, 46
- Kaziannis, S. 28
- Kelly, J. M. 29
- Kidd, A.K. 41, 42, 57
- King, R.B. 21, 22, 34
- Kingham, R.J. 14, 17
- Krushelnick, K. 19
- L**
- Lancaster, K.L. 8, 9, 41
- Landowski, B. 36
- Lewis, C.L.S. 8
- Li, B. 40
- Lintern, A. 43
- Livingstone, R. 23
- Long, C. 28
- Lopes, N. 8, 9
- Lowe, H.F. 45, 55
- Loosemore, H. 42
- Lukacs, A. 27
- Lysaght, M.A. 16
- M**
- Mackenzie, A. 38
- Magnusson, J. 30
- Malvestuto, M. 33
- Manton, J.C. 28
- Marangos, J. 23
- Marklund, M. 16
- Marshall, V. 36, 37
- Martin, J. 35
- Martin-Fernandez, M.L. 25, 38
- Mason, P. 43
- Matthewson, D. 42
- McCann, J.F. 22
- McGovern, D.M. 29
- McKenna, P. 7, 10, 11, 12, 46
- McKenzie, A. 25
- Meech, S.R. 27
- Minns, R. 23
- Mithen, J.P. 17
- Mohr, C. 27
- Moore, A.S. 9
- Morishita, T. 15
- Musgrave, I. 41
- N**
- Najmudin, Z. 8, 9, 10, 11, 12, 19
- Nakatsutsumi, M. 7
- Needham, S.R. 25, 38
- Neely, D. 10, 11, 12, 35, 37, 42
- Nemeth, G.R.A.J. 21, 22, 33, 34
- Neville, D. 46
- Newell, W.R. 21, 22
- Norreys, P. A. 8, 9, 19
- Notley, M.M. 8
- O**
- Olsson, S. 9
- O'Neill, P. 26
- Osterholz, J. 7, 46
- Osterrieder, A. 25
- P**
- Palmer, C.A.J. 10, 11, 12
- Parker, T. 51
- Parker, A.W. 24, 26, 28, 29, 39, 51
- Parmigiani, F. 33
- Pascu, S.I. 24
- Pasley, J. 14, 15
- Pattathil, R. 12, 35
- Pearson, A. 42
- Pepler, D.A. 41, 42
- Petersen, J.C. 23, 33
- Phillips, P.J. 43, 44
- Pickett, C.J. 28
- Poletto, L. 21, 33
- Pollard, M.R. 39
- Portius, P. 28
- Prasad, R. 10, 11
- Pryce, M.T. 28

Q

Quinn, S.J.	29
Quinn, K.E.	7, 10, 11, 46
Quinn, M.N.	7, 10

R

Rajeev, P. P.	35, 37
Ramakrishna, B.	7, 46
Ramis, R.	8
Rehman, A.	8, 9
Ressel, B.	33
Reynolds, D.	42
Reynolds, P.	26
Rice, P.	43, 46
Ridgers, C.P.	17
Rishworth, G.	42
Roberts, S.K.	25, 38
Robinson, D.	42
Robinson, D.A.	39, 41,
Robinson, A.P.L.	10, 11, 12, 14, 18
Rodríguez, R.	9
Rolfe, D.J.	25, 38
Romagnani, L.	7, 10, 11, 12
Römischi-Margl, W.	27
Rossall, A.K.	8
Ruddock, I.S.	31, 32
Russo, C.	8, 9

S

Santabarbara, S.	28
Sarri, G.	7, 8, 9, 46
Sawyer, C.	43
Schlenvoigt H-P.	7
Schreiber, J.	10, 11, 12
Scott, R.H.H.	8, 9
Seddon, E.A.	33
Serra, S.	45
Shaikh, W.	41
Sherlock, M.	17
Siano, M.	23
Silva, L.O.	19
Simons, J.P.	29, 30
Singh, D.K.	9
Sircombe, N.J.	18

Smith, R.A.	9, 37
Sparkes, I.	25
Spencer, C.L.	27
Spesvytsev, R.	23
Spindloe, C.	11, 44, 46, 55
Springate, E.	21, 22, 23, 33, 34
Stanca-Kaposta, E.C.	30
Stewart, A.I.	28
Strange, T.	40, 46
Streeter, M.J.V.	10, 11, 12, 37
Stuart, N.	41
Su, Z.	29, 30
Symes, D.R.	9, 35, 36, 37

T

Tallents, G.J.	8
Tanaka, K.A.	8
Tang, Y.	12
Tanimoto, T.	8
Tatarakis, M.	8, 9
Temporal, M.	8
Ter-Avetisyan, S.	10, 11, 12
Theobald, M.B.M.	24
Tolley, M.K.	45, 46, 55
Tolstikhin, O.I.	15
Tomlinson, S.	43
Toncian, T.	7
Tonge, P.J.	27
Townsend, D.	23
Towrie, M.	27, 28, 29, 39
Tresca, O.	7, 10, 11, 12
Trines, R.	8
Trines, R.M.G.M.	18, 19
Turcu, I.C.E.	21, 22, 23, 33, 34, 47
Tucker, R.W.	13
Tylee, A.	36, 38
Tynan, C.J.	25, 38
Tzoufras, M.	13, 19

U

Underwood, J.G.	23
-----------------	----

V

Van Der Hart, H.W.	16
Vassura, L.	7
Villoresi, P.	21, 33
Vlaic, S.	33
Volk, M.	30
Vorberger, J.	14, 20

W

Wagenaars, E.	8
Waghorn, P.A.	24
Wagner, B.	29
Ward, A.D.	25, 40
Ward, P.	42
Watanabe, S.	15
Webb, S.E.D.	25, 38
Weeks, M.	42
Wegner, U.	37
Weller, T.E.	31
White, S.	8
Willi, O.	7, 46
Williams, I.D.	21, 22
Wingfield-Stratford, J.	42
Winn, M.	25
Winstone, T.B.	41, 42, 57
Wojdyla, M.	29
Wright, J.A.	28
Wünsch, K.	20
Wyatt, D.	45
Wyborn, B.E.	42

X

Y

Yuan, X.	7, 10, 46
----------	-----------

Z

Zepf, M.	10, 11, 12
Zhao, R-K.	27

Plasma jets and narrow bandwidth ion spectra from thin foils irradiated at high laser intensity

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Abstract:

Evolution of collimated plasma jets and ion acceleration from micron to sub-micron thickness foils following the interaction of ultra intense (up to 3×10^{20} W/cm²) laser pulses is investigated. We characterized the correlation between laser intensity and target areal density for the formation of a supersonic dense plasma jet. With the maximum achievable intensity on target, jets with Mach number up to 10 have been observed from few microns thick Cu foils expanding longitudinally with a speed of 2×10^6 m/s. With circular polarization, the uniformity of jet density profile has improved significantly as compared to the jets observed from interaction of a linearly polarized laser. While the moderate energy (KeV) overdense plasma jets are formed efficiently for micron thickness target irradiated at 10^{20} W/cm² intensity, decreasing the target areal density or increasing irradiance on target leads to so-called light-sail RPA [1] regime in which the ions, independent of their charge to mass ratio, are accelerated to tens of MeV/nucleon energies.

Introduction: One of the main applications of high power lasers is to simulate various astrophysical phenomena in laboratories. Jets and collimated outflows are omnipresent in the universe associated with the formation of young stars, planetary nebulae, X-ray binaries and active galactic nuclei (AGNs) [2]. Although the experimental jets driven by high power lasers are of much smaller scales compared to stellar or extragalactic objects, they may provide an excellent opportunity to study in the laboratory the underlying mechanisms.

In laboratory experiments, plasma jets relevant to astrophysics have been generated typically by employing nsec long laser pulses of hundreds of joules energy [3-5]. In this Letter, we have characterised the conditions for the formation of supersonic jets driven by pico-second laser pulses of similar energies, as observed previously by Kar et. al [6]. At such high irradiance (around 10^{20} W/cm²), jets can be produced by the strong ponderomotive pressure exerted on thin targets by the laser. Via parametric scans (laser intensity, target areal density) we have identified the suitable conditions for the formation of a sustained supersonic jet. The temporal evolution of the jets up to ns time is studied by time resolved optical interferometry. Apparently, beyond the parameter ranges required for sustained plasma jet, strong modulation in MeV proton/ion spectra is observed as we increase the laser irradiance or decrease the target areal density. Preliminary analysis of the ion spectra will be discussed.

Experiment:

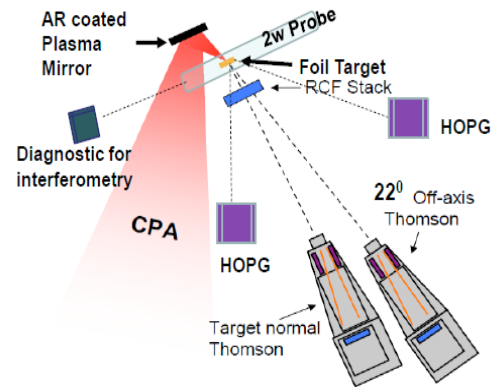


Fig 1. Schematic of the experimental set up.

The experiment was carried out in the Vulcan Nd: glass laser of Rutherford Appleton Laboratory, U.K., operating in chirped pulse amplification (CPA) mode. An $f=4.5$ ($f=3$) off-axis parabola was used to focus the laser down to a $10\text{ }\mu\text{m}$ full width at half maximum (FWHM) spot, attaining peak intensity of $3\times 10^{20}\text{ W/cm}^2$ on the target. A plasma mirror was used in order to suppress the intensity of amplified spontaneous emission and prepulses. The laser was irradiating the target at nearly 0 degree incidence angle. The polarisation of the laser on target was varied by using a quarter wave plate. The intensity on the target was varied, as required by the interaction intensity scans, by increasing the laser spot size on the target by translating the parabola along the focussing axis. Targets of various materials and thicknesses (free standing thin foils, $10\text{ }\mu\text{m}$ to 100 nm) were used in this experiment. The plasma outflow from the target was probed by employing a transverse Nomarsky interferometer [7] and shadowgraphy with high spatial (few microns) and temporal (ps) resolution. Figure 1 shows a schematic of the experimental arrangement. The probe beam was frequency doubled from a portion of the main CPA pulse to a wavelength 527 nm . The converted beam was splitted into two parts and projected onto the target, transverse to the interaction axis, from two different angles. The time of arrival of each beam on the target was controlled separately by delaying their path length. The setup was designed to achieve two ps snapshots of the interaction at different times in a single shot. The interferograms were recorded by 12 bit dynamic range CCD cameras. The plasma electron density profile from the interferogram was reconstructed by retrieving the phase map using fast Fourier transform of the fringe pattern in the interferogram, followed by Abel inversion of the phase map assuming a cylindrical symmetry [8].

The main diagnostic for multi Mev proton emission was Thomson Parabola Spectrometer (TP –Spec). Two high resolutions TP –Spec were fielded, one along the target normal direction and the other looking at 22° off axis to the target normal. Image plates (IP) were used to record the ion spectra. By using differential filtering the response of the image plate to different energy ions was calibrated which will be discussed in another report.

Result and Discussions: The temporal evolution of the collimated plasma jets at the target rear side was observed from 150 ps to 1.1 ns , after the arrival of the CPA on the target, over many shots. $1\text{ }\mu\text{m}$ thick Cu foil was used for this temporal scan and the laser intensity on target was kept at its maximum ($2\text{--}3\times 10^{20}\text{ W/cm}^2$). The plasma jets maintained their degree of collimation over ns time scales as shown in the fig. 2(a). From the temporal scan, the transverse and longitudinal dimensions of the jets were inferred at different probing time, as shown in the Fig. 2(b). Over this probing time period, the results suggest a non-linear (a power law) time dependent velocity for the longitudinal expansion of the jets, whereas an almost constant expansion velocity is observed for the transverse direction. Consequently, the Mach number of the jets, estimated as a ratio between the longitudinal to transverse velocities, has a strong time dependence during the first few hundreds of ps after the interaction.

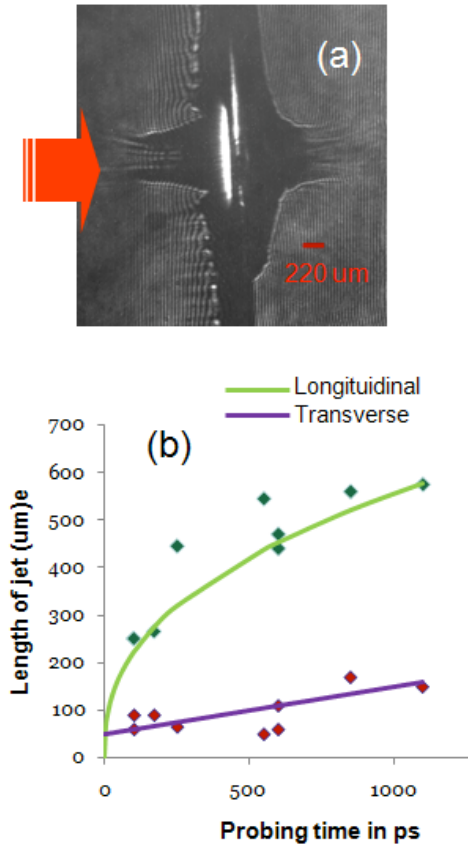


Fig 2. (a) Plasma jet at 850 ps after the interaction with $1\text{ }\mu\text{m}$ Cu target. (b) Temporal evolution of the plasma jets produced from $1\text{ }\mu\text{m}$ thick Cu target irradiated at $2\text{--}3\times 10^{20}\text{ W/cm}^2$.

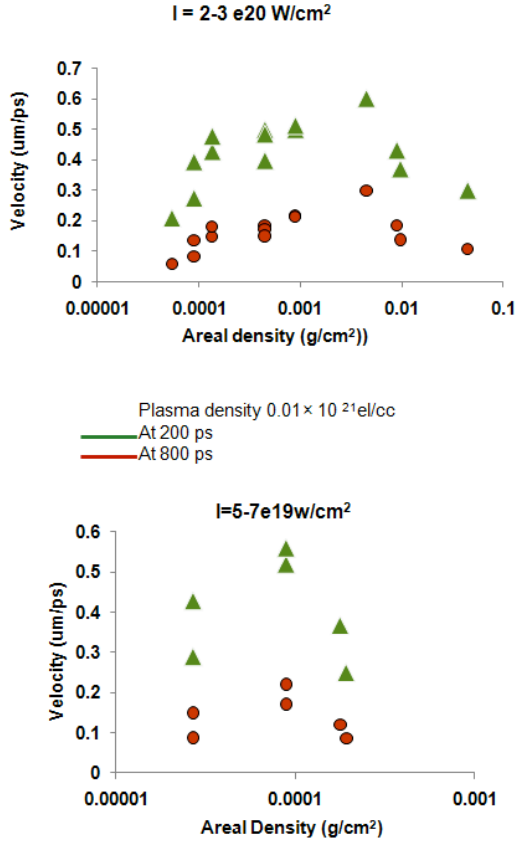


Fig 3(a) and (b). Velocity of jet for different target areal densities (at given laser intensity)

In order to parametrise the underlying mechanism behind the collimated jet formation, scans were made observing the rear side plasma jets at different laser intensity for same target and at same intensity for different target areal density (thickness $\times$ density). For a given laser intensity, the jet velocity increases as we decrease the areal density (moving to thinner targets or lower Z targets) and attains maximum at a certain value of areal density. As we decrease the areal density further, the jet velocity drops sharply. In fact for the lower areal density targets the interferograms shows plasma density profiles, which one would expect from a blow-off target. However in this regime, the ion spectrum shows features which suggest the onset of the so called light sail radiation pressure acceleration mechanism [1], which will be discussed later. A similar trend of jet velocity variation with areal density is observed for two different laser intensities and the areal density for which the jet velocity is maximum is lower, as expected, for lower irradiance [see fig 3(a) and (b)]. This behaviour is also observed for circular polarisation of the interaction laser. As shown in the Fig,

4(b) a sustained high mach number plasma jet from thinner targets can be produced by lowering the laser intensity commensurately. For example, a factor of 10 less intense laser produces jet of similar mach number from 100 nm target than that required in case of 1 μm thick target.

However, there is a distinct difference between the plasma jets produced from a laser at linearly polarised to circularly polarised. It can be seen pedagogically by comparing the raw images of the jets shown in Fig. 2(a) and 4(a) that the plasma front of the jets in case of circular polarisation (Fig. 4(a)) is flat and uncorrupted as compared to the one in linear polarisation (Fig. 2(a)).

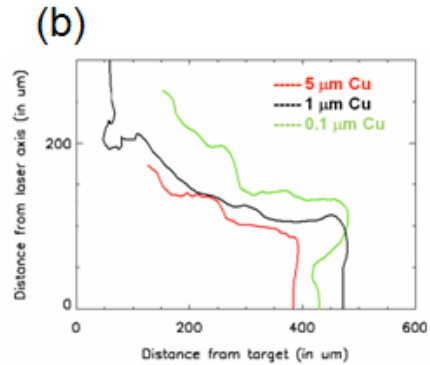
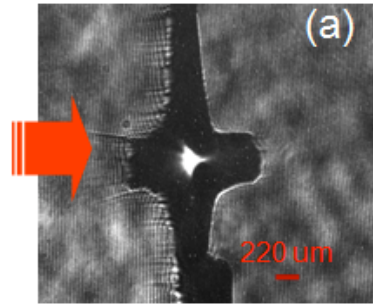


Fig 4(a) Plasma jet at 600 ps after the interaction with $1 \mu\text{m}$ Cu target at circular polarisation of laser. (b) Thickness scan for $1 \mu\text{m}$ target at circular polarisation of laser for the density of 10^{19} el/cc , for $0.1 \mu\text{m}$ target the laser intensity was comparatively lower than other two targets.

Increasing the laser intensity on target, or by lowering the target areal density beyond the values required for the formation of a sustained plasma jets, the MeV proton and ion energy spectra observed by thompson parabolas

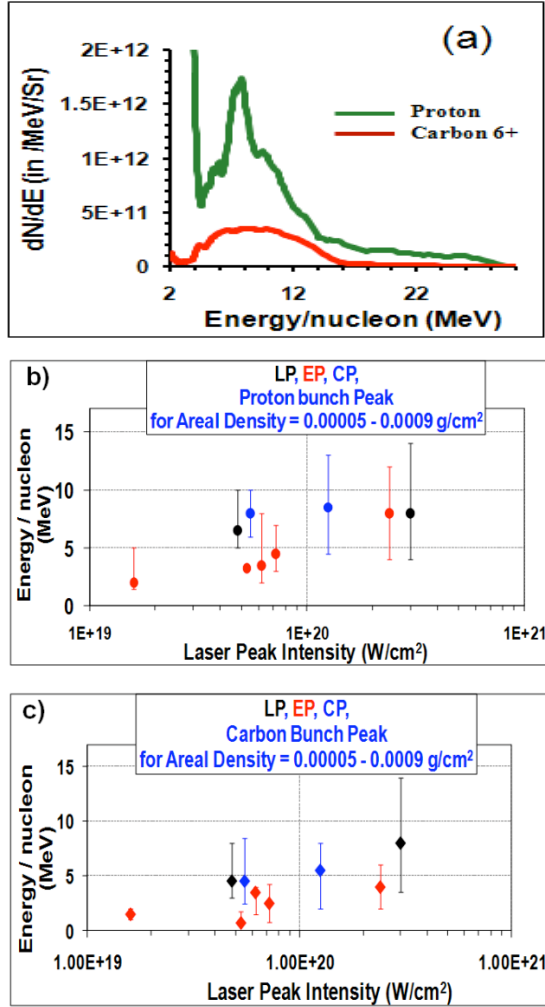


Fig. 5: (a) Ion spectra obtained from the interaction of 100 nm target at 3×10^{20} W/cm 2 intensity. (b) Peak energy of proton and (c) carbon bunch at areal density of target 0.00005- 0.0009 g/cm 2 for different laser intensity. Here data points show the peak energy of bunch and error bars show the width of the bunch

become pronouncely modulated. Instead of a quasi maxwellian energy spectra for protons and carbon ions observed in the case of stable plasma jet formation regime, the spectra contains a peaked feature towards the higher energy side (fig. 5(a)). Moreover, the peaked feature in both carbon and proton spectrum lies almost close to each other over the energy/nucleon axis. Further decreasing in the target areal density or increase in laser intensity lead the bunched feature in barbon and proton spectra shifting towards higher energy (fig. 5 (b) and (c)). Priliminary 1D particle in siumulation with multiple target layers (bulk target layer with thin contaminat layers on the both sides) suggest the peaked feature in the

spectra originates from the radiation pressure acceleration of contamination layer present at the laser side of the target, whereas, the exponetial low energy part in the spectrum orinates from TNSA acceleration of the target rear surface contaminant layer.

Conclusion: The dynamics and characterization of highly collimated supersonic plasma jets produced from thin targets irradiated by ultra-intense laser pulses is reported. Preliminary analysis suggests a role of the radiation pressure of the intense laser beam in the jet formation. In case of stronger drive on mass density leads the plasma jets loosing its significance and acceleration of narrow band proton and carbon spectra upto 10s of MeV per nucleon.

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Influence of overlapping high-intensity laser beams on electron and ion generation and transport

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Introduction

Laser facilities that will explore the Fast Ignitor (FI) concept [1] are planned to comprise many bundled laser beams to produce the electron or ion beams required to achieve ignition of the pre-compressed core. For example, present planning for the HIPER FI facility [2] includes up to 42 bundled beams to reach the energy levels required for ignition. Similarly, (i) the FIREX-I laser facility in Japan, a test-bed for a planned full FI facility, will be composed of 4 bundled beams and (ii) the Omega EP facility is composed of two kJ, ps beams [3]. Up to now, most of theoretical and experimental studies of FI [4] have been performed using a single beam. For example, studies of the electron transport of GA currents using hybrid codes [5] assume that a single beam of electrons is generated. This is also the case for laser-cone FI studies, see e.g. Ref. [6]. Having multiple beams overlapping on the target could however significantly modify the end result of the interaction. Regarding electron-driven FI, the overlapping of the beams at the critical density interface could impact the generation and the potentially unstable propagation of the ultra-high current electron beams, in which the greatest uncertainties of FI still lie. Regarding ion-driven FI [7], the mixing of the hot electron sheaths emerging from the solid could be detrimental, modifying the ion acceleration process and creating spatial modulations in the global sheath that lead to unwanted caustics in the ion beam [8]. All these issues involving bundled beams need to be clarified for the FI studies to progress quantitatively. We therefore performed an experiment using two different setups addressing two topics in this context. In the first, we used two temporally separated co-linear laser beams to study the influence of having a delay between them on the plasma interaction. In the second, we used two spatially separated, quasi-orthogonal, beams to study the influence of a spatial and temporal separation between, on the plasma interaction. All these configurations were used irradiating solid (metal) targets.

Experiment

The experiment was conducted on the Vulcan TAW laser facility at the Rutherford Appleton Laboratory. This facility provided up to two independent high-energy laser pulses at the fundamental frequency (i.e. of 1053.5 nm central wavelength with a 2 nm band width). After stretching and

amplification, the main Vulcan laser pulse was split into these two individual pulses (CPA#7 and CPA#8). Each laser pulse contained approximately 50 J of energy (after compression, with a pulse duration down to 1.2 ps) and was focused onto the targets using, for each, an f/3 off-axis parabolic mirror (OAP) (Fig. 1 (a)). The focal spot for each was measured to be $6.5 \mu\text{m} \pm 2 \mu\text{m}$ (full width half maximum) and $\sim 35\%$ of the energy was contained between the peak of the spot and $1/e$ of that peak (Fig. 1(b)). The corresponding peak intensity on target was thus $\sim 2 \times 10^{19} \text{ W/cm}^2$. To detect the ions and electrons resulting for the laser interaction, we employed stacked detectors as shown in Fig. 2(a) and magnetic spectrometers, equipped with imaging plates (IPs) as detectors, placed in the forward and backward direction (with respect to the laser irradiation direction), along the target normal. For protons, we used stacked calibrated radiochromic films (RCF) (of HD-810 and MD-V2-55 type). They detected the laser-accelerated multi-MeV protons that are electrostatically accelerated by the laser-produced energetic electrons [9]. The stack was placed $\sim 35 \text{ mm}$ behind the target foil in the normal direction and a hole was drilled through to let the particles reach freely the rear side magnetic spectrometer.

RCFs measured the spatial distribution of proton beam at discrete energies corresponding to the proton deposited energy in the form of Bragg peaks. As also shown in Fig.2(a), for the electrons, behind the RCFs, we positioned a stack of 5 IPs separated by 1.5 mm thick Al plates (type TR2025) to resolve in

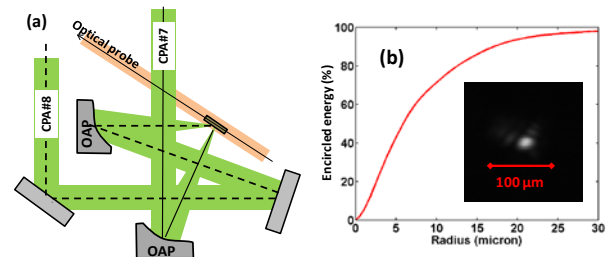


Fig. 1 (a) General layout of the experiment and CPA laser beams configuration. (b) Azimuthally averaged radial profile of a measured focal spot. (inset) Image of the focal spot.

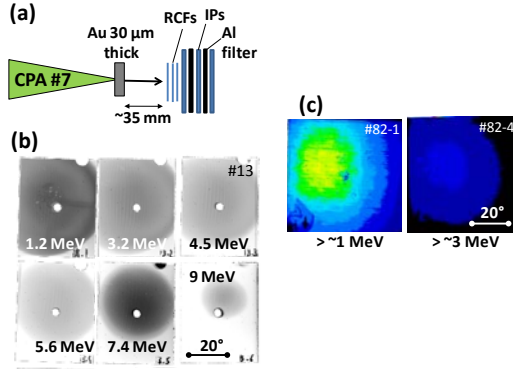


Fig. 2 (a) Setup for ions and electrons detection. (b) Typical proton beam distribution images obtained with RCFs. (c) Spatial distribution image of $>1\text{ MeV}$ and $>3\text{ MeV}$ electrons detected by the stack of imaging plates (IPs) shown in (a).

energy and angle the hot electrons [10].

In order to have a basis of comparison when we used a dual beam irradiation configuration, we first assessed the result of a single beam irradiation on target. Fig. 2(b) shows typical RCF images of the proton beam obtained in the configuration when a single laser beam interacted with a $30\text{ }\mu\text{m}$ thick Au foil. The typical maximum proton energy was $8 - 10\text{ MeV}$. The divergence of the 7 MeV proton beam was around 30 degrees full angle. For the same shot, the measure of the fast electron beam divergence is shown in Fig. 2(c). The color bar here represents a logarithmic scale (QL value). In addition to these diagnostics, to assess the level of preplasma in front of the target at the time of the main laser pulse irradiation due to the laser pedestal, the plasma expansion profile of the laser-irradiated targets was diagnosed using an optical probe that was passed transversely across the target surface (as shown in Fig. 3(a) and Fig. 1(a)). The probe beam was picked up from a small part of the CPA#7 that was then frequency-doubled to 2ω (527 nm). The target images were obtained with 16 bit Andor technology CCD camera (type: DX420-BN, pixel-size: $26\text{ }\mu\text{m}$), using a 10 times magnification. To minimize the plasma self-emission signal, (I) an iris (opened $\sim\phi 1\text{ mm}$) was placed in the imaging line at the first probe beam focal point using a $f = 950\text{ mm}$ collecting lens. Then, (II) the signal was filtered with a ω fundamental frequency-cut filter (BG38) as well as a 2ω band pass filter (CVI Melles Griot: F10-527-4-2) that were put in front of the CCD. A typical image taken by this diagnostic for the same shot as Fig. 2(b) is shown in Fig. 3(b). The probe was timed to cross the target 10 ps after the main interaction pulse hit it. The image shows a large plasma expansion on the laser-irradiated surface of the target with extent of approximately $410\text{ }\mu\text{m}$ (transverse) and $220\text{ }\mu\text{m}$ (longitudinal). We observe that, contrary to the target front, the target rear does not exhibit any expansion, which is consistent with the high-energy proton acceleration that is observed [11]. The reference image taken just before the shot is also shown in Fig. 3(c).

The experimental setup of the first experiment investigating the energetic electron beam propagation when using two temporally separated collinear laser pulses is shown in Fig. 3(a). To achieve such irradiation configuration, the CPA#7 was spatially divided in two half-beams that could have a temporal delay between them but that were nonetheless focused on same location on the same interaction target surface ($20\text{ }\mu\text{m}$ Au foils) as they both used the same focusing OAP. The relative timing of the two separated beams was adjusted by altering the path delays with $\sim 1\text{ ps}$ precision using an interferometric technique. As illustrated in Fig. 3(a), to diagnose

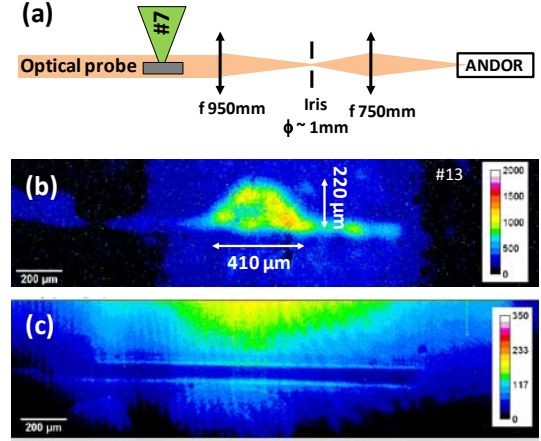


Fig. 3. (a) Setup for preplasma expansion (from the target front-surface) measurement using the optical probe beam. (b) The image corresponds to the same shot as in Fig. 2(b). It is recorded 10 ps after the interaction pulse irradiation. (c) A reference image is taken before each shot.

the result of this interaction, we used a probing proton beam that acted as a transverse charged particle probe for the fields associated with the electrons emerging from the rear of the interaction target [12]. To produce this probing beam, we used the CPA#8 that was focused onto a $25\text{ }\mu\text{m}$ thick gold foil to generate the adequate protons. The interaction targets were bent in order to minimize the deflections due to global target charge-up which would have otherwise prevented sampling of the accelerating fields. The probing proton virtual source size was small enough, so that the images can have excellent spatial resolution ($\sim$ a few μm s). The multilayer arrangement of the RCF stack detector, providing a spectral resolution, allows a temporal multiframe capability. The magnification of the arrangement is given by $M \sim (L + 1)/l$, where $l = 3 - 4\text{ mm}$ was the distance between the proton target and the interaction area, while $L \sim 35\text{ mm}$ was the distance between the interaction target and the RCFs. This gives M equal to $8 - 10$ in our experimental conditions. This technique is mainly sensitive to field gradients, which are detected via proton density modulations in the probe beam cross section. Figure 3(b) shows typical proton probing data obtained when the CPA#7 interaction beam was full and not split in two halves. Darker regions correspond to higher proton densities in the proton beam cross section. Each frame corresponds to a different probing time. The piling up of the probe protons with bubble-like structure appearing at later times is due to the sudden drop of the electric field ahead of the acceleration front. Such structures are similar to the ones described in [12]. When we divided temporally the CPA#7 pulse into two, the expansion velocity of the sheath was slightly decreased compare to the full beam configuration. This corresponds to the expected reduction of focal intensity of each laser by reduction of energy by half, as well as the increase of the focal spot size. The latter is because of the f-number doubling linked to the near field size reduction. However, one striking difference compared to the single laser interaction is that only in the split beam configuration did we observe, shortly after the interaction, a jet-like proton deflection pattern. Such jet had an extent of $> 500\text{ }\mu\text{m}$, and were observed reproducibly when there was a small delay (1.5 and 3 ps) between the two beams. One should note that the fast plasma expansion front from the interaction target cannot reach such a far region ($> 500\text{ }\mu\text{m}$) during such a short time ($< 5\text{ ps}$ after laser irradiation). A detailed analysis is still under way.

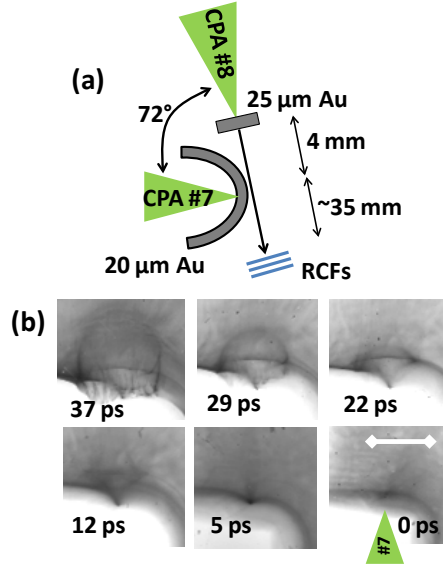


Fig. 4 (a) Experimental setup for investigating the energetic electron beam propagation when using two temporally separated collinear laser pulses. (b) Typical proton probing data obtained when the CPA#7 interaction beam was full and not split in two halves. The scale bar corresponds to 500 μm in the interaction target plane. The time 0 corresponds the time when the CPA#7 arrives at the interaction target.

Figure 5 shows the other experimental setup we used to investigate the energetic electron beam propagation when using two laser pulses irradiating the targets with a large angular difference. CPA#7 and #8 then irradiated the target surface with a 72 degrees angular difference. The laser beams were synchronized such that they arrived at the surface of the gold 30 μm thick target simultaneously. In this setup, we changed the spatial separation between the laser beams over a series of shots such that the mutual separation between the laser-accelerated energetic electron beams created by each laser beam and arriving at the rear surface was comprised between -45 μm (i.e. when the lasers overlap at the target front) to $\sim +150$ μm . Here, as a first approximation, we assumed that the major part of electrons propagate geometrically straight along the laser axis [13]. The proton beams accelerated from the target were recorded on a stack of RCFs as well as dispersed by the magnetic spectrometer. One can expect that the sheath electrostatic fields, which have an extent of typically 100 μm [14], would overlap or partially overlap at the target rear (depending on δx) and influence the proton acceleration. Indeed, when the sheath overlapped constructively (i.e. for $|\delta x| \leq 50$ μm), we observed a clear enhancement of the accelerated proton energy by a factor of ~ 1.5 compared to the energy obtained in the single beam configuration. Such enhancement disappeared when we spatially separated the beams too much, as expected. Another striking feature was that we observed, in the dual beam interaction, on the RCFs a strongly collimated energetic proton beams on top of the usual diverging proton beams (see Fig. 5). As observed in all previous experiments using single-beam irradiation, the diverging proton beams showed a decrease of their divergence angle with proton energy [15], as is also seen in Fig. 1(a). Such a variation of divergence angle is expected for a transversally bell-shaped electron density distribution [14]: the highest energy protons are accelerated in the central, high-density portion of the sheath, whereas lower energies come from the wings of the sheath distribution and thus are emitted at larger angles. In contrast, a strongly collimated beam appeared in the case of dual overlapping beams. It exhibited an almost constant divergence angle ($\sim 17^\circ$ full angle) when varying the proton energy. Such structure is observed repeatedly when δd is sufficiently small

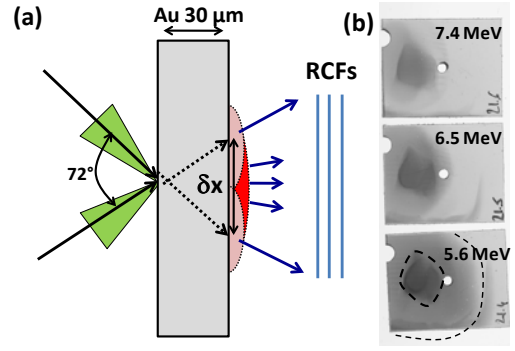


Fig. 5 (a) Experimental setup for investigating the energetic electron beam propagation when using two laser pulses irradiating the target with a large angular difference. We varied the spatial separation δx . (b) Proton distribution images obtained when the lasers overlap at the target front.

(i.e. $|\delta d| \leq 100 \mu\text{m}$). Such collimated beam could be attributed to the overlapping of bell-shaped electrostatic sheaths. This would result in creating a plateau in the electron density profile between the two electrostatic sheaths at the target rear surface created by each laser pulse, as illustrated in Fig. 5.

Acknowledgements

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Characterization of a line focus back-lighter

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Introduction

Opacity measurements at EUV wavelengths of high energy density plasmas are difficult to undertake as the probe has to overcome the high self emission of the opaque plasma and it is difficult to create sufficiently uniform and well characterized plasma for accurate measurements. Laser produced EUV and x-ray back-lighters have been used previously to perform opacity measurements [1,2] when the flux of x-rays generated is sufficient to 'outshine' the plasma self emission. More recently, much brighter plasma based EUV lasers have been employed to probe an iron plasma [3]. In the present experiment, we set out to use a short duration (3ps) Ne-like Ge laser at 19.6nm to probe the opacity of laser heated iron.

Information regarding opacity is relevant to a number of different fields including solar and astrophysical modelling [4], inertial confinement fusion [5,6], and free-electron laser experiments [7] and there exist a number of different computer codes for simulating opacity [8,9,10,11,12]. In order to test the accuracy of these codes and solve discrepancies between them, experimental benchmark data is required.

This experiment ultimately generated an EUV back-lighter in a set-up similar to that used to create a plasma based x-ray laser, without lasing being observed. A line focus back-lighter can produce bright emission over a broad spectral range as the more intense spectral lines approach black-body intensities due to the high optical depth along the plasma line. This emission comes from a small area and can be pointed at a sample. This paper is primarily concerned with the characterisation of the back-lighter with the aim of improving our understanding of the line focus EUV back-lighter and to investigate why lasing was not observed. Accurate information regarding the plasma conditions within the EUV source is required for accurate opacity measurements. Transmission measurements through iron plasma from this experiment using EUV radiation are reported elsewhere [13].

Experiment

The back-lighter was created using two optical pulses from the VULCAN Nd:Glass laser system at the Rutherford Appleton Laboratory to irradiate a germanium target of thickness 500nm deposited onto a glass slab of 6mm width. A pre-pulse of duration 300 ps and energy of 20J was focused into a line 100 μ m x 6mm, using a refracting lens and a spherical mirror, giving an irradiance of $\sim 8 \times 10^{12}$ W cm $^{-2}$. A main (CPA) pulse of 3ps duration had an energy of 35J and was focused into a line of similar dimensions using a single f/3 off-axis parabola, giving an irradiance of $\sim 1 \times 10^{15}$ W cm $^{-2}$. The line foci of both beams were overlapped on the target, with a delay between the pulses of $\Delta t=400$ ps.

A 50nm thick iron foil, tamped with 100nm thick parylene-N (C $_8$ H $_8$) on both sides, was heated using another CPA beam from the VULCAN laser and the plasma created probed with the EUV back-lighter. The second CPA beam contained 25J

energy in a duration of 1ps and was focused to a spot size of about 200 μ m in diameter on the opacity target. The delay between the heating pulse and the main CPA pulse was varied between 5 and 20 ps to allow probing at different stages of iron plasma evolution. Probing results have been presented elsewhere [13].

In order to characterise the line focus Ge back-lighter, a crossed-slit camera with an Andor CCD was positioned at an angle of 22 degrees with respect to the plane of the target normal. A front 15 μ m horizontal slit provided a vertical magnification of 3.56 with the back 100 μ m vertical slit giving a horizontal magnification of 1.20. The vertical and horizontal resolution was 19 μ m and 180 μ m respectively. The crossed-slit camera was filtered using 3 μ m aluminium and 25 μ m beryllium so that emission in the range 0.5 to 5 keV was recorded. The CCD had an exposure time of 500ms so we record time integrated Ge emission over the laser plasma lifetime.

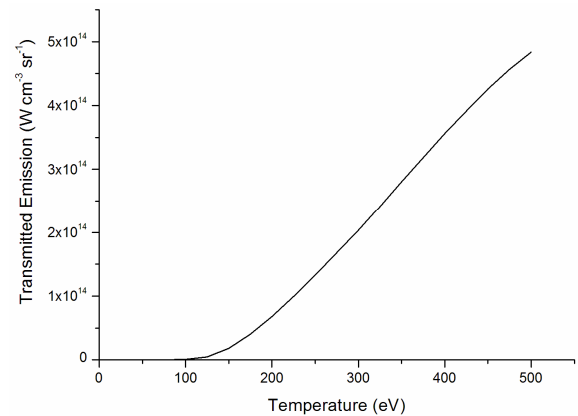


Figure 1. Spectrally integrated emission transmitted through filters of 3 μ m Al and 25 μ m Be as a function of temperature for Ge as determined with the FLYCHK code [14] and using filter transmission data from Henke et al.[15]. The electron density assumed is that seen in the region of highest gain as predicted by Ehybrid (5x10 21 cm $^{-3}$).

Germanium Back-lighter Characterisation

The crossed-slit images provide an 'emission map' of the Ge back-lighter and have been characterized using simulated data from the spectral modelling code FLYCHK [14]. The code was used to evaluate total emission from the Ge plasma as a function of photon energy and temperature for the density in the region of highest gain and emission as predicted by the Ehybrid code [16] ($n_e = 5 \times 10^{21}$ cm $^{-3}$). Multiplying the photon energy dependant transmission [15] of the filter with the total emission and integrating over frequency gives the total emission as seen by the crossed-slit camera per unit volume (see figure 1). For our filter combination, the relationship between the transmitted emission and temperature is approximately linear in the

temperature range of 150 – 500eV, with a threshold for any significant emission at $\approx 120\text{eV}$.

We assume that the plasma emission through the crossed-slit filter combination ($3\mu\text{m}$ Al, $25\mu\text{m}$ Be) is dominated both spatially and temporally by the hottest temperature and highest density ($5 \times 10^{21} \text{ cm}^{-3}$) plasma. The area of plasma viewed by each pixel on the CCD is calculated using the geometry of the crossed-slit camera. A thickness of emitting plasma can be determined by calculating the plasma expansion from the turning point density by calculating the ion sound speed, v_s , for each temperature and considering a characteristic length scale, $v_s \Delta t$ (where $\Delta t = 400\text{ps}$, is the time between the pre-pulse and the main pulse). The time of emission of the plasma (100ps) is found from simulations using the 1D hydrodynamic code Ehybrid [16] (figure 2).

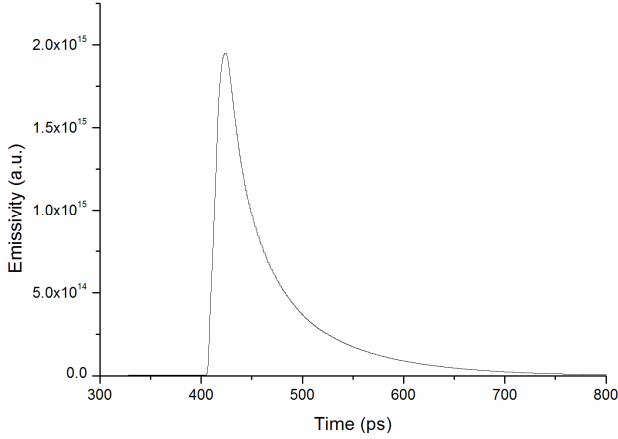


Figure 2. Emission of radiation from a Ge plasma transmitted through the filter combination for our crossed-slit camera as a function of time calculated by the Ehybrid [16] code.

A calibration factor for the CCD (8eV/count) is used to deduce the absolute number of counts per pixel associated with the transmitted emissivity. Figure 1 then enables the crossed-slit images of recorded emission to be converted to a spatial variation of electron temperature (figure 3, 4). This conversion of CCD counts to temperature is reasonably accurate ($\pm 10\%$) due to the rapid linear increase of emission with temperature and the threshold temperature for any significant emission (see figure 1). Our error estimate for the temperature measurement of $\pm 10\%$ does not include the error associated with our assumption of a spatially and temporally constant density and temperature. The incident laser intensity profile has a width of $100\mu\text{m}$ and comparing this with the deduced temperature profile width of $150\mu\text{m}$, the relationship $T_e \propto I^\beta$, results in $\beta = 0.44 \pm 0.05$ in agreement with temperature scaling results [17].

Simulations using Ehybrid demonstrate peak ionisation levels in agreement with the FLYCHK model used to determine the temperature profile of the back-lighter. The peak temperature for the pre-pulse only back-lighter was found to be 120eV or less. A peak temperature of $155 \pm 20\text{eV}$ is found when the two pulse set-up is used. These temperatures explain the presence of spectral lines from Ge XX to GeXXIII [18,19,20,21,22] as seen in the spectrum shown in figure 5 and the significant change in emission between the pre-pulse and double pulse plasma. The figure 5 spectrum was recorded with a flat field spectrometer viewing down the line focus axis.

Although the experimental set-up was similar to that used to generate a plasma based x-ray laser, no x-ray lasing was observed and one of the motives of the present characterization of the back-lighter was to investigate why x-ray lasing was not seen. A temperature analysis of the pre-pulse only shots shows the temperature to be $< 120\text{eV}$, and according to ionisation curves simulated using the collisional-radiative code FLYCHK,

the plasma then consists mainly of Ge^{19+} or lower ionisation stages. This ionisation is consistent with the peak ionisation determined via simulations using Ehybrid. For lasing the main pulse has to couple enough energy into the plasma to raise the ionisation level up to the Ne-like ionisation stage (Ge^{22+}) and provide the population inversion necessary for lasing, within the 3ps pulse length.

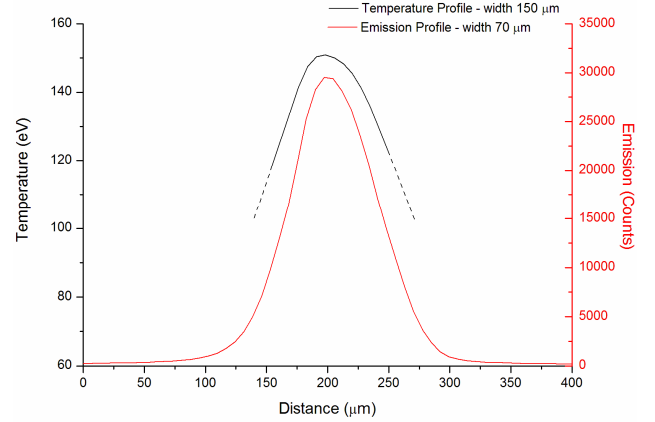


Figure 3. Emission cross-section over the line profile and the resulting temperature profile deduced as discussed in the text.

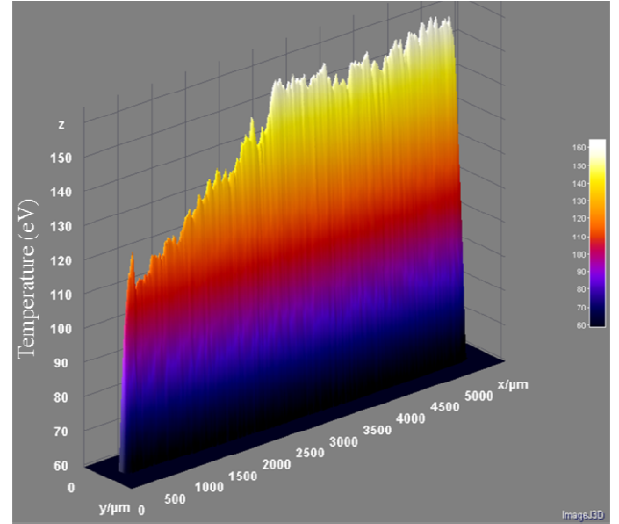


Figure 4. 2D temperature profile of the Ge back-lighter.

Due to the short pulse duration (3ps) of the laser, the main pulse energy is largely coupled at higher densities, meaning any x-ray laser photons produced may be refracted out of the gain medium, reducing amplification. This effect has been reported previously by Smith et al. [23] and so has been investigated through simulations with Ehybrid (see figure 6).

Ehybrid demonstrates that the experiment produces the maximum gain within the Ge plasma at a density of $5 \times 10^{21} \text{ cm}^{-3}$. The region of gain as simulated by Ehybrid has width $\Delta s \approx 4\mu\text{m}$ (figure 6) and has a peak gain value of $G=200 \text{ cm}^{-1}$. Refraction at such high densities is significant. We can write that the refraction angle θ is related to the density scale length Δx and electron density n_e by [24]

$$\theta \approx \frac{L}{\Delta x} \frac{n_e}{2n_c}$$

where n_c is the x-ray laser critical density. Setting the maximum refraction angle $\theta = \Delta s/L$, gives that the x-ray laser propagation distance L in the gain region is such that

$$L \leq \sqrt{\Delta s \Delta x \frac{2n_c}{n_e}}$$

For $\Delta s = 4\mu\text{m}$, $n_e = 5 \times 10^{21} \text{ cm}^{-3}$ and density gradient as shown in figure 6, we obtain $L \leq 100 \mu\text{m}$, implying maximum gain length product $GL \leq 2$ which would not be observable.

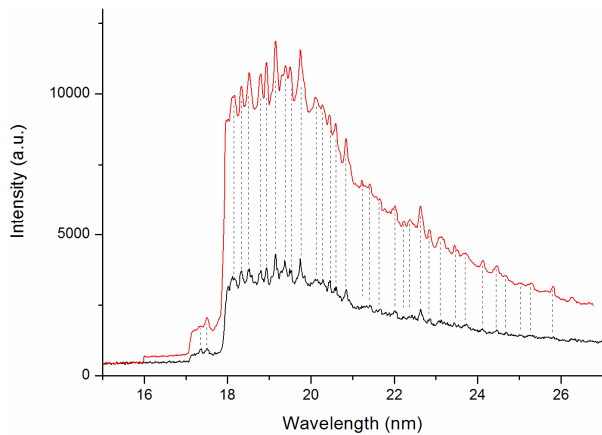


Figure 5. Ge spectrum as recorded by a flat field spectrometer. The red line demonstrates the spectrum originating from a back-lighter created using the two-pulse setup and the black line shows the spectrum from a back-lighter created using only the pre-pulse. The drop in intensity at 18nm is due to the CCD chip being partially covered with aluminium. The intensity drop at 17nm is the aluminium K edge.

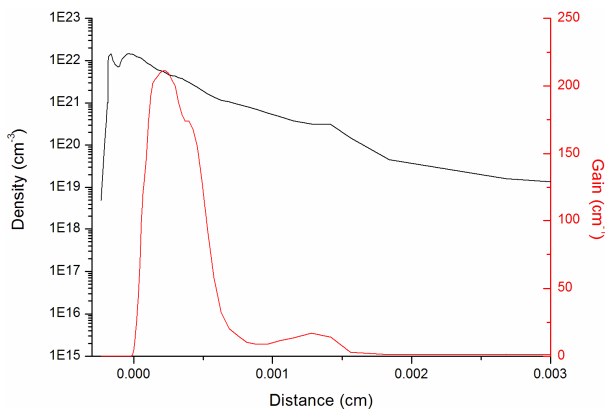


Figure 6. Density and small signal gain profile at the time of peak gain, $t=409\text{ps}$, 6ps after the end of the main CPA pulse as simulated by Ehybrid for the conditions of the experiment.

Conclusions

This experiment has demonstrated how a plasma back-lighter can be well characterized through imaging the time and spectrally integrated emission profile of the plasma using a crossed-slit camera. By applying a detailed model of emission to the back-lighter images, a spatially dependant electron temperature profile can be inferred and used as a useful diagnostic of the plasma. Further study has shown the pulse duration of the main CPA pulse not to be ideal for lasing. Simulations using Ehybrid demonstrate that the short pulse duration and other conditions of the experiment result in energy coupling at higher densities where the increased refraction deflects lasing photons out of the gain region inhibiting amplification.

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Creation of persistent, straight, 2mm long laser-driven channels in underdense plasmas for Fast Ignition applications

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Abstract

The experimental study of the behavior of deuterium plasma with densities between 2×10^{18} and $2 \times 10^{20} \text{cm}^{-3}$, subjected to a 6 TW, 30 ps, $3 \times 10^{18} \text{Wcm}^{-2}$ laser pulse, is presented. Conclusive experimental proof is provided that a single straight channel is generated when the laser pulse interacts only with the lowest densities. This channel shows no small-scale longitudinal density modulations, extends up to 2 mm in length and persists for up to 150 ps after the peak of the interaction. Bifurcation of the channel after 1 mm propagation distance is observed for the first time. For higher density interactions, above the relativistic self-focusing threshold, bubble-like structures are observed to form at late times. These observations have implications for both laser wakefield accelerators and fast ignition inertial fusion studies.

Introduction

There are a number of applications of intense laser pulses that require the formation of a stable channel in an underdense plasma [1, 2]. These include quality control of multi-MeV electron bunches generated in laser wakefield accelerators [3–5], particularly the reproducibility and selection of the bunch energy [6], as well as high brightness X-ray generation in these channels by betatron oscillations [7] and fast ignition inertial confinement fusion (FI) [8]. In the latter case, Tabak et al. [8] proposed to add a hundred of picoseconds scale pedestal to the ignition pulse, in order to generate a channel through the underdense plasma and to push the critical surface close to the pre-compressed fuel. Such a channel would then allow the ignition pulse to propagate practically undisturbed through the mm-scale coronal plasma surrounding the dense fuel. This scheme has several advantages compared to the alternative cone-guided concept which has already demonstrated, in small-scale experiments, efficient coupling between

petawatt pulses and the core [9]. Firstly, it promises less complex target manufacture, allowing low-cost mass production for inertial fusion energy purposes. Secondly, it allows targets with a higher degree of spherical symmetry to be compressed.

In the past few years, channeling driven by high intensity laser pulses has been extensively studied in a number of experiments (e.g. [10–13]) mainly by optical interferometric techniques, as well as X-UV radiography [14]. These experiments concerned either short duration laser pulses (≤ 1 ps) or relatively small observation distances (≤ 0.2 mm). All these interactions, however, employed relativistic laser pulses; in this regime, detrimental instabilities such as longitudinal modulations induced by relativistic self-focusing [13], long wavelength hosing [13] and self modulation [10] have been seen to occur.

Li et al. [15] have shown, with the aid of 2D Particle-In-Cell (PIC) simulations, that stable channels can be generated when a pulse of sufficiently long duration is used; the sustained ponderomotive pressure eventually wins over the instabilities. These simulations were run with regard to a plasma density in the range $0.1n_c \geq n_e \geq 1.02n_c$. However, a directly driven, ignition scale implosion will generate a large region of less dense plasma ahead of the compressed fuel.

Therefore, as part of the HiPER project [16], an experiment was conducted on the Vulcan laser facility at the Rutherford Appleton Laboratory [17] whose specific aim was to study laser channeling in underdense plasma. The experiment used a laser pulse, emulating the one proposed by Li et al. [15], focused onto a supersonic deuterium gasjet with variable density. Here, the results of this experiment are presented. It is demonstrated that, when the power of the laser pulse is below the threshold for relativistic self-focusing, the sole

ponderomotive force is able to create a longitudinally smooth channel extending for the whole gasjet extension (i.e. 2 mm) and persisting for up to 150ps. The channel formation has been reproduced in matching 2D PIC simulations of this experiment.

Experimental setup and raw data

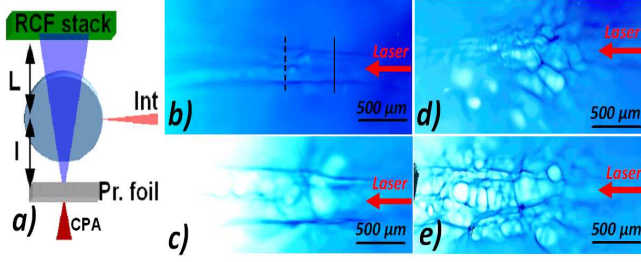


Figure 1: **a.** Schematic top-view of the experimental setup. **b.** $n_e = 2 \times 10^{-3}n_c$, $t_{prob} = 100$ ps. **c.** $n_e = 2 \times 10^{-3}n_c$, $t_{prob} = 150$ ps. **d.** $n_e = 2 \times 10^{-1}n_c$, $t_{prob} = 100$ ps. **e.** $n_e = 2 \times 10^{-2}n_c$, $t_{prob} = 100$ ps. All the radiographs refer to a probing proton energy of 4 MeV

The gasjet backing pressures chosen were to be in the range 1 - 100 bars corresponding, once fully ionised, to $n_e = 2 \times 10^{18}\text{cm}^{-3} - 2 \times 10^{20}\text{cm}^{-3}$ (i.e. $2 \times 10^{-3} - 2 \times 10^{-1}n_c$). The laser parameter were as follows: $\tau_L = 30$ ps, $P_L = 6$ TW, $I\lambda^2 = 3 \times 10^{18}\text{Wcm}^{-2}\mu\text{m}^{-2}$. The laser power was sufficient to study conditions below and above the threshold for relativistic self-focusing, given by $P_{cr} = 17(n_c/n_e)\text{GW}$ [18]. The interaction was probed in the transverse direction via the proton radiography technique [19] by the use of a high fidelity proton beam created during the interaction of a second laser beam (pulse duration 1 ps, energy $E = 60$ J and intensity 10^{19}Wcm^{-2}) with a $20\mu\text{m}$ thick metallic foil (see Fig.1.a for a sketch of the setup). The proton beam, after having probed the plasma, was recorded on a stack of dosimetrically calibrated RadioChromic Films (RCF), [20] geometrically magnified by a factor of $M \approx L/l \approx 6$ (Fig. 1.a). The proton probe was timed to traverse the plasma at $t_{prob} = 100 - 150$ ps after the peak of the 30-ps duration interaction pulse.

Typical data showing the features observed by proton imaging for $n_e = 2 \times 10^{-3}n_c$ are displayed in Figs. 1.b and 1.c. Fig. 1.b shows that, at 100 ps after the peak of the interaction, a channel, $300\mu\text{m}$ wide, is present with sharp edges for the entire length of the gas jet itself and no longitudinal modulations can be seen within the channel, in contrast to previous experimental observations [10, 11, 13]. The channels starts to deteriorate only after 150 ps (Fig. 1.c).

For densities laying above the threshold for relativistic self-focussing, persistent bubble-like structures were observed both outside and within the channel (see Fig. 1.d and 1.e). In this regime, the channel does not survive unperturbed during the time when the proton beam traversed the plasma at late times.

An interesting feature arising directly from the observation of the raw data for the smooth channel is its apparent bifurcation in the center of the gas jet in Fig. 1.b.

Discussion

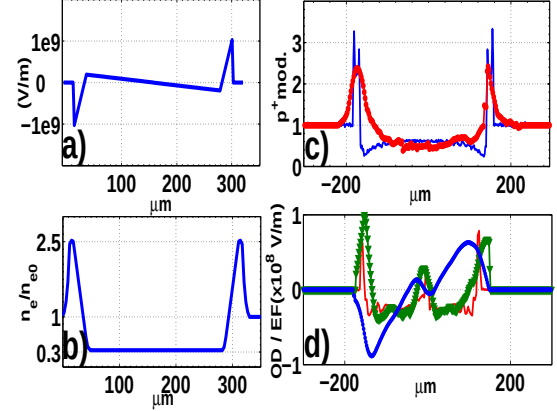


Figure 2: **a.** 1D profile of the electric field distribution used as the input for the PT simulations. **b.** Related electron density normalized to $2 \times 10^{18}\text{cm}^{-3}$. **c.** Red line: experimental proton modulation across the channel (solid line in Fig.1.b). Blue line: PT simulated proton density modulation. **d.** Green triangles: experimental optical density modulation across the bifurcated region (dashed line in Fig.1.b). Red line: optical density from the related PT simulation. Blue line: experimental electric field distribution.

In order to estimate the electron density modulation across the channel associated with the proton modulation recorded by the RCFs, Particle Tracing (PT) simulations of the propagation of the probing proton beam through the plasma have been performed for both the bifurcated and single channel regions. This code tracks proton trajectories of specified energies (4 MeV in the present work) from a point-like source through a time-dependent 3D electric field distribution up to the proton detector. This provides a 2D proton density mapping at the detector plane. For the case of the single channel region, the simulation is only able to reproduce the relevant features of its radiographic image provided that an electric field, symmetric along the laser propagation axis, (shown in Fig 2.a with the related electron density distribution shown in Fig 2.b) is assumed. This is consistent with a piling up of the plasma density up to 2-3 times the background density

at the channel walls and an inner density depletion of $\approx 70\%$. A comparison between the measured proton probe beam modulation at the RCF plane (along the lineout highlighted in Fig. 1.b) and the simulation is shown in Fig. 2.c, revealing close agreement. For the bifurcated channel, the code reproduces the features visible in Fig. 1.b only if two different cylindrically symmetric sub-channels, similar in shape to the single channel, are assumed (see Fig 2.d for a comparison between the experimental and simulated optical density results). This confirms that the channel does split into two cylindrically symmetric sub-channels.

Two-dimensional particle-in-cell (PIC) simula-

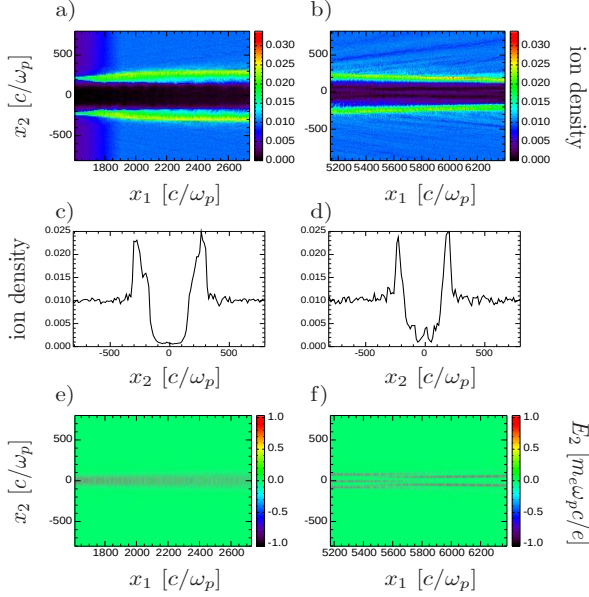


Figure 3: Simulated ion density and laser electric field distribution for both the single (images **a.** and **e.**) and bifurcated regions (images **b.** and **f.**). Images **c.** and **d.** show transverse crosscuts of images **a.** and **b.** respectively.

tions have been carried out using the code OSIRIS [21]. A fully ionised deuteron-electron plasma has been assumed; its density profile is exponential with $50 \mu\text{m}$ scalelength until it reaches 1% of the critical density, and constant after that. Laser parameters are as follows: amplitude $a_0 = 1.0$, corresponding to 10^{18} Wcm^{-2} , $10 \mu\text{m}$ spot diameter, 20 ps FWHM duration; the laser is polarised along the x_2 axis.

Simulation reveals that a plasma channel is formed with steep walls (2-2.5 times the background density n_b) and a low density near its axis (0.1-0.2 times n_b). This is similar to the experimental results, apart from the widening of the experimental channel and the dulling of its walls, brought about by its evolution in time (100 ps versus 16 ps). It has to be noted that a slightly higher initial density is needed as a simulation parameter in order to match the experimental data basically due to geometrical reasons: a 2D geometry in fact confines less the laser. We repeated in fact the simulation at a lower density of $0.002n_c$, matching the density used in the experiments more closely. The channel formation process was basically identical, but the self-focusing was less prominent, leading to a creation of 4 rather than 2 parallel beamlets.

Conclusions

In conclusion, the experimental study of channeling driven by a 6 TW, 30 ps laser pulse through an underdense deuterium plasma has been presented. Below the regime of relativistic self-focussing, the sole classical ponderomotive force is able to drill a stable and smooth channel, ideal for subsequent pulse guiding through the large regions of tenuous plasma in a direct drive fast ignition scenario. Multi-dimensional PIC and particle tracing simulations of the laser-driven channel formation show good agreement with the experimental observations.

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Observation of post-soliton expansion following laser propagation through an underdense plasma

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Abstract

The expansion of electromagnetic post-solitons emerging from the interaction of an intense laser pulse with an underdense plasma has been observed up to 100 ps after the pulse propagation, when large numbers of post-solitons were seen to remain in the plasma. The temporal evolution of the post-solitons has been accurately characterized with a high spatial and temporal resolution. The observed expansion is compared to analytical models and three dimensional particle-in-cell results providing indication of the polarisation dependence of the post-soliton dynamics.

The experimental characterization of such structures is of fundamental importance for high intensity laser-plasma physics studies since they are the major responsible for the dissipation of the laser energy.

Introduction

During its propagation through an underdense plasma, the laser experiences a significant energy loss. Being this energy loss fully adiabatic, it is mostly translated into a red shift of the laser light [6]. In the case of initial plasma densities close to the critical density, this frequency decrease may eventually lead the laser to locally experience an overcritical plasma, thus being trapped in plasma cavities. These cavities, whose radius is of the order of the electron collisionless skin depth ($l_e = c/\omega_{pe}$, where ω_{pe} is the Langmuir plasma frequency) are usually referred as electromagnetic (e.m.) sub-cycle solitons [4]. These structures tend to be accelerated along plasma density gradients [3, 7] and, therefore, are slowly propagating, if not steady, in a homogeneous plasma. For times longer than the ion plasma period, the Coulomb repulsion of the ions left inside the cavity causes it to radially expand, and the soliton nature is lost: such late-time evolution of a

soliton is thus commonly referred as a *post-soliton* [8]. This mechanism is similar to the Coulomb explosion that occurs inside laser channels following relativistic self-focussing [1].

Post-soliton expansion has been analytically studied [8, 9], using the so-called snowplow model [10] and the isolated spherical resonator model [11], and numerically, using Particle-In-Cell (PIC) codes [8]. Experimental observation of post-soliton structures was first reported in [12], where soliton remnants were observed in the dense region of a plasma resulting from the laser-driven explosion of a thin foil. Due to the nature of the plasma employed, clouds of bubble-like structures were detected thus preventing a precise characterisation of the temporal evolution of the post-solitons.

Here, we report the first experimental observation of well isolated post-soliton structures. This allows their evolution to be resolved and followed over a significant temporal window. The experimental results are compared to analytical and three dimensional (3D) PIC code models.

Experimental setup and raw data

The experiment was carried out at the Rutherford Appleton Laboratory employing the VULCAN Nd:glass laser operating in the chirped pulse amplification mode [13]. A sketch of the set up is given in Fig. 1: 200 J of 1 μm laser light contained in a 30 ps Full Width Half Maximum pulse (“Int” in Fig. 1) were focussed to a peak intensity of $3 \times 10^{18} \text{ W cm}^{-2}$ at the edge of a supersonic deuterium gas jet with a backing pressure ranging from 1 to 100 bar. This resulted, once fully ionized, in an electron density of $10^{18}\text{--}10^{20} \text{ cm}^{-3}$, which is 0.001–0.1 times the non-relativistic critical density n_c . The interaction was diagnosed via the proton radiography technique [12, 14], which uses, as a particle probe, a laser accelerated proton beam, arising

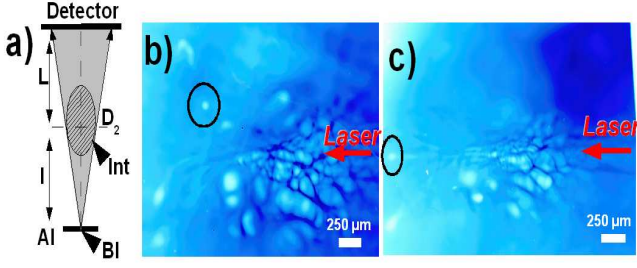


Figure 1: **a)** Top view of the experimental arrangement. **b)**, **c)** Radiographs of two different shots: outlined with dark circles are the density bubbles interpreted to be post-solitons.

from the interaction of a secondary laser pulse ($\tau \approx 1$ ps, $E \approx 100$ J, $I \geq 10^{19}$ W cm $^{-2}$, “BI” in Fig. 1) with a 20 μm thick aluminium foil. The virtual point-like source [15] allows imaging of the interaction area with a geometrical magnification $M \approx (l + L)/l \approx 6$, where $l \approx 6$ mm and $L \approx 3$ cm, see Fig. 1.a. The probe beam, after having passed through the gas jet, was recorded by a stack of RadioChromic Films (RCF) [16].

The data set comprised about 30 shots in which both the deuterium density and the probe time were varied. Two typical proton radiographs obtained at a density of $0.1n_c$ are shown in Fig. 1. These images were obtained with protons of energy ≈ 4 MeV, 100 ps after the beginning of the interaction. As a rule of thumb, the electric fields are directed from the regions of lighter blue color (reduced proton flux) towards the regions of darker blue color (increased flux). In both images, a channel created by the laser pulse is visible in the low density region at the edge of the gas jet. In the dense region inside the gas jet a strongly modulated deflection pattern is visible along the laser propagation axis. This scaly region highlights the presence of a cloud of bubbles that appear merged or overlying one another in this 2D projection, possibly surrounding the laser-driven channel. Such a region visually resembles the cloud of solitons that was experimentally and numerically observed in [12]. Ahead of and around this region, isolated bubble-like structures are visible (black circles in Fig. 1), most of them located at the end of laser filaments, as numerically predicted in [3]. These bubbles are associated with strong probe proton depletion with sharp edges. We note that bubble-like structures were never seen at electron densities of $0.01n_c$ or less. We ascribe these bubbles to post-solitons.

Discussion

Considering the isolated bubbles allows following the fundamental properties of the post-solitons temporal evolution. Thanks to the multi-frame capability of proton radiography [14], it is possible to follow their temporal evolution of these bubbles in the range 80–130

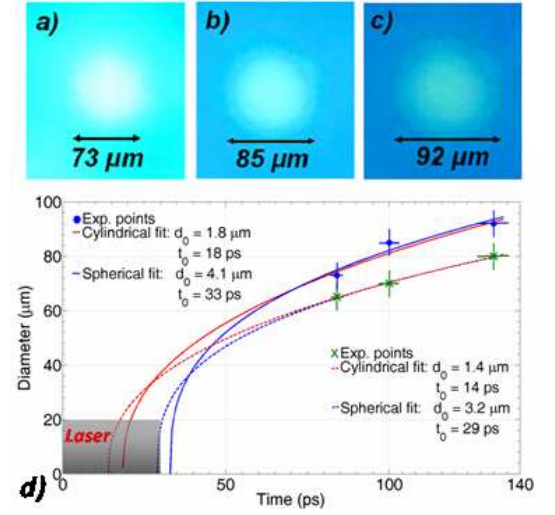


Figure 2: Zoom in on the bubble structure outlined in Fig. 1 in different layers of the RCF stack corresponding to 84 ps (**a**), 100 ps (**b**) and 132 ps (**c**). **d**) Bubble diameter as a function of time and fits using results from the snowplow model for cylindrical and spherical post-solitons.

ps after the beginning of the interaction, i.e. 40–90 ps after the laser has left the gas jet. Both bubbles analyzed were found to be effectively stationary in the laboratory reference frame and to expand preserving a roughly circular shape (Fig. 2).

Bulanov and Pegoraro [9] give analytical results for the expansion of 1D planar, 2D cylindrical and 3D spherical post-solitons using the snowplow model. In 3D the diameter of the sphere is given by $d_0(3^{3/2}t/t_s)^{1/3}$ for $t \gg t_s$, where t_s is given by $\sqrt{2\pi d_0^2 n_0 m_i / \langle E_0^2 \rangle}$, d_0 is the initial diameter, n_0 is the initial ion density, m_i is the ion mass and $\langle E_0^2 \rangle$ is the time average of the square of the initial oscillating electric field inside the post-soliton. We fitted the experimental results with this function taking the initial diameter d_0 and the time of creation of the soliton t_0 as free parameters and $\langle E_0^2 \rangle^{1/2} = 2 \times 10^{12}$ V m $^{-1}$, which is roughly the average value of 40% of the initial laser field. Even though this function was able to fit the experimental data (see Fig. 2), it implied a creation time at the end of, if not after, the laser pulse duration, which is not physically sensible. We therefore tried the 2D cylindrical result $d_0(5t/t_s)^{2/5}$: this gave a more physically meaningful fit with a creation time close to the peak of the laser pulse in both cases. The initial diameters from the cylindrical fits are also more reasonable than those from the spherical fits, being $\approx 1 \mu\text{m}$ instead of $\approx 3 \mu\text{m}$, since we have $c/\omega_{pe} \approx 0.53 \mu\text{m}$ (see Fig. 2 for the fits result). Other bubbles in Fig. 1 and in different shots (not shown) have been found to expand in a similar fashion.

In order to understand why a 3D structure follows predictions for 2 rather than 3 dimensions, we carried out a 3D run with the PIC code OSIRIS [17]. We considered a linearly polarized laser pulse with a wavelength of $1\ \mu\text{m}$, Gaussian spatial and temporal profiles with FWHM of $6\ \mu\text{m}$ and $1\ \text{ps}$, respectively, and a peak intensity of $3 \times 10^{18}\ \text{W cm}^{-2}$ incident on a fully ionized deuterium plasma with a density of $0.1n_c$. The interaction was followed for $5\ \text{ps}$. The simulation box was $350 \times 50 \times 50\ \mu\text{m}$, divided into 2.4×10^8 cells each having 2 particles for electrons and 2 for deuterium ions, the time step was $0.196\ \text{fs}$. A number of longer 2D runs, with a larger number of particles per cell and a range of plasma densities, were also carried out for s -polarization (laser electric field out of the plane) and p -polarization (laser electric field in the plane). The ion

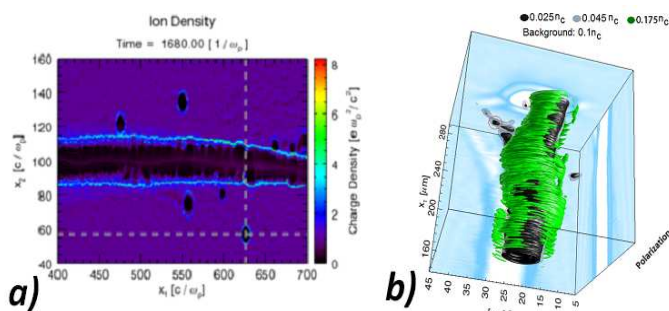


Figure 3: **a)** Ion density at 3ps from 2D PIC modeling for s -polarized light. **b)** Iso-surfaces of the ion density at 4.11 ps from 3D PIC modeling. Sub-channels are formed in the s -plane (perpendicular to the laser electric field) following leakage of the laser from the channel formed. Several post-solitons are located in these sub-channels.

density in Fig. 3.b shows prolate spheroid post-solitons,

with an aspect ratio of 5:3, lying outside the channel formed by the laser in the plane perpendicular to the laser electric field. In the 2D runs solitons were only ever formed for s -polarized light (see, for instance Fig. 3.a). This dependence of soliton creation upon the laser polarization is in line with the PIC code results reported in [3]. The solitons were formed as the result of laser leakage due to its breaking-up in filaments that led to the creation of sub-channels departing from the main channel (as visible in both Figs 3.a and 3.b).

The non-spherical shape of the post-solitons gives a first indication as to why the spherical scaling may not apply. The observed polarization dependence of soliton formation suggests why the spherical scaling does not apply. The snowplow model assumes in fact total reflection of the trapped light at the overcritical soliton wall and therefore no plasma heating. This is a good approximation only for s -polarized light, since a p -polarized wave will be indeed absorbed by the overcritical walls [18]. This might explain why p -polarized light is not able to excite stable e.m. solitons [3].

Conclusions

In conclusion, we have reported the first experimental measurements of the late time expansion of post-solitons following the propagation of a relativistically intense laser pulse through an underdense plasma. The post-soliton expansion has been temporally resolved with high temporal and spatial resolution. The post-soliton expansion is best described by the analytical prediction for cylindrical, not spherical, post-solitons. 3D and 2D PIC code results suggest that this is due to polarization effects.

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Perspectives on radiative blast waves in laser-heated clustered gases

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Introduction

The continuous growth in scale of laser systems has enabled the study of material behaviour under increasingly extreme conditions. In the 1990's this capability led to the inception of *laboratory astrophysics*, in which researchers perform well characterised experiments and use hydrodynamic scaling arguments to explain and predict what occurs in astrophysical phenomena^[1-4]. The experiments also provide important data for verification and validation of various aspects of numerical codes such as atomic physics, equation of state, radiative transfer and hydrodynamics. Many of these investigations have been performed with very large facilities such as the Omega laser and the Z z-pinch because of the large laser energies or x-ray fluxes required to drive materials into regimes of interest. Laboratory astrophysics studies are core objectives for the NIF and HiPER fusion projects and could also be applied to Space Science projects. In this report we discuss an alternative method to create a high energy density environment which takes advantage of the remarkable absorption of short pulse laser light exhibited by clustered gases^[5-7]. This allows us to conduct laboratory astrophysics experiments with smaller scale chirped pulse amplification lasers^[5-18].

Regimes of radiative shock

One particular area of interest is that of *radiative shocks*, which are common in astrophysics. These cause complicated structures, such as those seen in supernova remnants, because the polytropic index is lowered through radiation and ionisation making the gas more compressible and so more susceptible to instabilities. For a strong shock the compression can be related to the polytropic index using the energy conservation jump condition to be $c = (\gamma + 1)/(\gamma - 1)$. This gives an ideal gas limit of $c=4$ for $\gamma=5/3$ and $c=7$ for $\gamma=4/3$, the value for a radiation dominated gas. In general, the calculation of a shock compression is more complicated because the terms for ionization and radiation energy flux in the jump condition cannot be ignored^[19].

The ultimate goal for experiments is to drive shocks into the *radiation dominated regime* when the radiation energy density (or pressure) exceeds the thermal energy density (or pressure). More realistically the shocks fall into the *radiative flux regime* so that radiative effects are evident but the pressure is still dominated by the thermal contribution. This is shown in Fig. 1 where these thresholds are shown by the red and cyan lines^[2]. For the shock to be affected the plasma must be optically thin to the radiation emitted by the shocked material and also the timescale for this loss must be faster than the time taken for convective heat transport. This gives us two additional limits in Fig. 1^[1], that the photon mean free path is larger than the scale size of the system (green), and that the radiative cooling time is shorter than the ratio of sound speed to scale size (blue).

These conditions are easily met in low density astrophysical environments such as supernova remnants^[1], but it is challenging to reach such a regime in the laboratory. We show

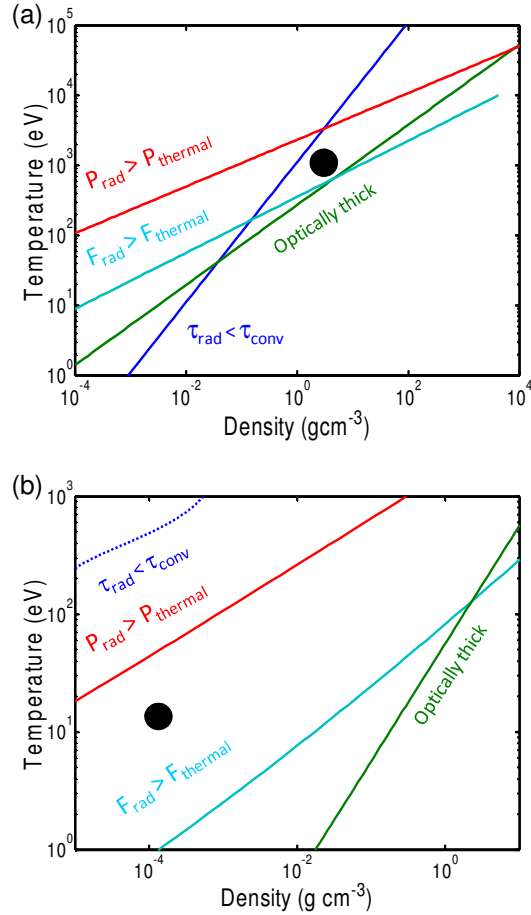


Figure 1. Plot representing the transitions between the various shock regimes described in the text. Cyan and red lines indicate the temperatures at which the radiative flux (F_{rad}) and pressure (P_{rad}) become dominant over the material quantities. Green and blue lines indicate when the photon mean free path is larger than the characteristic size of the system and the radiative cooling time (τ_{rad}) is shorter than the gas convection time (τ_{conv}) including only bremsstrahlung. In (b) the condition indicated by the dashed blue line is calculated using a radiative cooling function. The black circles mark the approximate properties of shocks created in (a) plastic and (b) clustered gases.

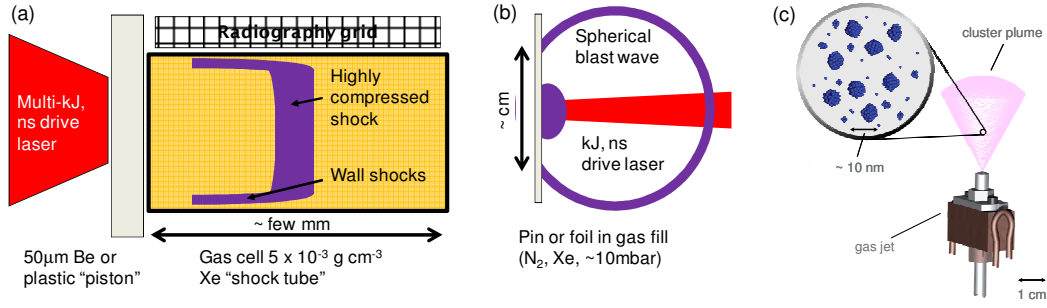


Figure 2. Methods to create radiative shocks (a) Piston driven shock, (b) solid target blast wave, (c) cluster blast wave.

this in Fig. 1(a) which corresponds to a typical experiment in a solid or foam target (represented by fully ionised carbon with a scale size $h=100\mu\text{m}$). Here we have followed the method of Ryutov *et al.*^[1] and Drake^[2] and used the blackbody expression for radiative flux and bremsstrahlung calculations for the radiation mean free path and cooling rate, which do not account for line radiation. The desired regime is tightly constrained and the high temperatures necessary mean that the shock speed must be very high ($\sim 100\text{kms}^{-1}$). This is the reason that these experiments require kJ drive energies and correspondingly large laser systems. We can alleviate this requirement by launching shocks in gases. Because the density is so much lower ($\sim 10^{-4}\text{gcm}^{-3}$), the shock speed needed to enter the radiative flux regime reduces to $\sim 10\text{kms}^{-1}$.

For the partially ionised gas, line radiation is the dominant energy transport mechanism meaning that the approximations used for the solid target are poor ones. To correctly categorize cluster shocks involves a complicated calculation of the radiative properties of the plasma considering line emission and without any assumption of equilibrium. We have recently started to apply the RAPCAL code^[21] to the problem and the resulting calculation will be of great use to the laboratory astrophysics community. Until this is complete, we consider the plot shown in Fig. 1(b) for partially ionised xenon ($Z^* = 0.63 (T_e v)^{1/2}$ ^[2]). The green, cyan and red lines are still calculated as in Fig. 1(a) so may not be accurate. For the blue line, we have used the (astrophysical) cooling rate from Post *et al.*^[20] which is more appropriate than using the bremsstrahlung rate but may still be incorrect for the densities used in laboratory experiments. Nevertheless, the rapid cooling of the gas suggests that a wide range of densities and temperatures will fall into the radiative flux regime.

Creating radiative shocks

Three methods have been used to create radiative shocks in the laboratory (See Fig. 2). The first is to make a “shock tube”^[22] by driving a solid density plastic or beryllium piston into a xenon gas cell. These experiments have been conducted at Omega^[3,23] and LULI^[3,24,25] using kJ lasers at a moderate intensity of $\sim 10^{14}\text{Wcm}^{-2}$ in a large smooth focal spot ($\sim 500\mu\text{m}$ diameter) to create a planar geometry. Because the laser pulse is long ($\sim \text{ns}$) there is a continuous injection of energy to the shock, creating a *stationary* structure. Shock speeds are very high ($>50\text{kms}^{-1}$) and because of the strong radiative losses the shocked gas is highly compressed ($c>30$).

A different type of shock is created following a sudden release of energy in a zero-extension, instantaneous explosion. In this case, a shock moves into the surrounding medium sweeping up material into a thin dense shell called a *blast wave*. The shock decelerates and decays as it expands and, if the shocked material is hot, radiative losses cause the collapse of the shell to very high density and the onset of thin shell instabilities^[26-29]. This is the situation encountered in supernova remnants, which are some of the most spectacular objects we observe. Radiative

blast waves can be generated experimentally by using a kJ laser to irradiate a pin or foil within a moderate to high Z background gas^[3,30,31]. The high laser energy is required to maintain a strong shock over relatively large spatial scales ($\sim \text{few mm}$). Alternatively the laser energy can be deposited directly into the gas to launch shocks^[5-18,32,33]. Ordinarily a gas absorbs only a small fraction of the laser light, but if clusters form then the absorption is dramatically increased and a hot ($T_e \sim \text{keV}$), high energy density plasma (up to 10^8Jcm^{-3}) is created. This plasma filament subsequently explodes into the ambient gas quickly forming a cylindrical blast wave^[5]. In this way high Mach number shocks can be launched using high intensity ($>10^{17}\text{Wcm}^{-2}$) lasers with energies as low as 100mJ provided that the pulse duration is short enough to couple energy into clusters before they explode (sub-picosecond timescale).

Radiative shocks can be categorized according to the optical depth of the gas ahead of (upstream) and behind (downstream) the shock. This is important to determine the suitability of a laboratory experiment for astrophysical scaling^[2] as shown in Fig. 3. Shocks in a supernova evolve from the top-right to the bottom-left of this plot. Initially, in the core of the supernova, both regions are optically thick. As the shock breaks out at the surface the precursor becomes optically thin and later, as the explosion forms a supernova remnant, the postshock region also becomes optically thin. The Omega and LULI experiments performed with solid density pistons fall into the ‘thick-thin’ regime. The low density gas blast wave experiments are expected to be ‘thin-thin’, although Drake suggests that because of the large volume of gas ahead of the shock, these conditions could actually be ‘thin-thick’. From our calculations we estimate the photon mean free path to be very similar to the scale size of the cluster blast waves so this differentiation is not so well defined.

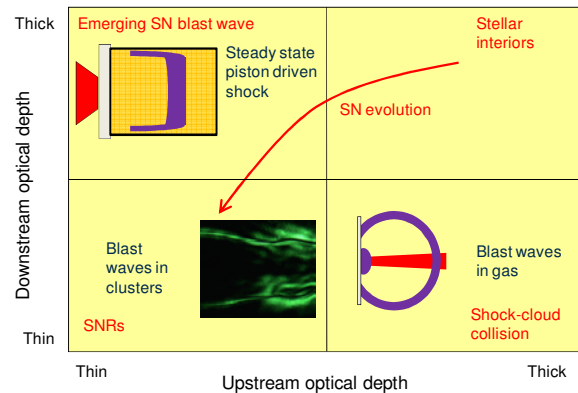


Figure 3. Categorising radiative shocks by upstream and downstream optical depth. Adapted from Ref. 2.

Characterising cluster blast waves

In order to assess the suitability of cluster shocks for scaling to astrophysical conditions we must be able to accurately define our experimental conditions. We have previously made some estimates of the plasma properties^[13] and here we further consider the morphology of our blast waves. The structure of a typical blast wave launched in xenon clusters with a laser energy of 350mJ is shown in Fig. 4. This is taken at 17ns when the material has been swept up into a thin shell behind a steep shock front. Ahead of this, a large precursor with a peak electron density about half of the post-shock value extends almost 1000 μ m into the ambient gas. This precursor is less prominent with lower laser energies and lower gas densities. Additionally, when the gas density is decreased the shock decelerates more quickly and the thickness of the shock shell is reduced. These three effects are consistent with a lowering of the opacity of the gas, meaning that less energy is deposited in the precursor and there is a higher energy loss from the system. A measurement of the shock front trajectory in low density xenon (radius $\sim t^{0.39}$) suggests that the blast waves are fully radiative i.e. 100% of the incoming kinetic energy is radiated away^[10].

From the electron density measurement shown in Fig. 4, we can attain some properties of the plasma. The ambient density in this case was $\rho = 0.25 \text{ kgm}^{-3}$ ($10^{18} \text{ atoms cm}^{-3}$) so in the precursor we have a direct measurement of the average ionization $\langle Z \rangle \sim 6$. From the shell thickness, and assuming all the material lies within this shell, we obtain a compression of ~ 2 . Using the postshock electron density of $14 \times 10^{18} \text{ cm}^{-3}$ we find that the postshock $\langle Z \rangle$ is similar at ~ 6 . If we assume that the plasma is in local thermal equilibrium (LTE) we can then estimate the pre- and post-shock temperature using $\langle Z \rangle$ and ρ to be $\sim 8\text{eV}$. The constant temperature across the shock front implies that we have driven a shock in the *supercritical* (SC) regime. This is feasible since the threshold velocity for a SC shock at this relatively low density is $\sim 7\text{kms}^{-1}$ ^[34], less than our measured shock velocity of $\sim 10\text{kms}^{-1}$. Further evidence comes from the shape of the precursor on some of our shots which have a plateau in the electron density profile which is commonly associated with SC shock formation. However, the assumption of LTE is a poor one since the plasma is not optically thick to the radiation.

The low compression we observe is a consequence of the high temperature of the precursor, which results in the pre-shock Mach number being drastically reduced ($M \sim 1.6$) and so negating the strong shock approximation. In fact a calculation of the compression is further complicated because the gas is partially ionized, affecting the polytropic index. The energy conservation jump condition across the shock can be generalized to take into account both the ionization and the radiation flux^[19]. However, the calculation of these fluxes is very complicated because the plasma is non-LTE and dominated by line emission. In addition, energy is not necessarily conserved across the shock because both pre- and post shock regions radiate energy in all directions.

To address these issues sophisticated numerical codes are necessary. There has been some success comparing experimental data to simulation results from hydrocodes which use the LTE approximation^[8-10,17]. Edwards *et al.*^[9] matched the trajectory of a xenon blast wave and identified the switch between a *radiative phase*, lasting from shock formation until several tens of nanoseconds, and an *energy recovery phase* when the blast wave sweeps up energy it has previously deposited in the precursor. We have observed this transition, at about the same time in the shock evolution, in several experiments since then^[17]. We find reasonable agreement of simulation results with the shock shell electron density profile (Fig 4(a), Ref. 17) but the code overestimates the length of the precursor. Keilty *et al.*^[10] compared the radiative cooling rates

given by a non-LTE calculation by Post *et al.*^[20] and those given by the LTE HYADES hydrocode^[35] and found a large discrepancy for xenon. This highlights the need for a non-LTE approach to the simulations but as yet we have been unable to apply non-LTE versions of the hydrocodes.

Recently we have started investigations using the RAPCAL code^[21]. This code can calculate radiative properties for optically thin and thick plasmas for both low and high Z over a wide range of density and temperature. This will enable us to more accurately determine gas opacities and radiative cooling rates without the assumption of LTE. Initial calculations were performed for a xenon plasma at a density of $2 \times 10^{-4} \text{ gcm}^{-3}$ and a temperature of 10eV which is similar to our experimental conditions. From Table 1 it would appear that the plasma is in LTE, but this is a consequence of using the LTE temperature value as an initial condition, which is a crude approximation. We are currently working to resolve this by using direct measurements to create the inputs for simulations.

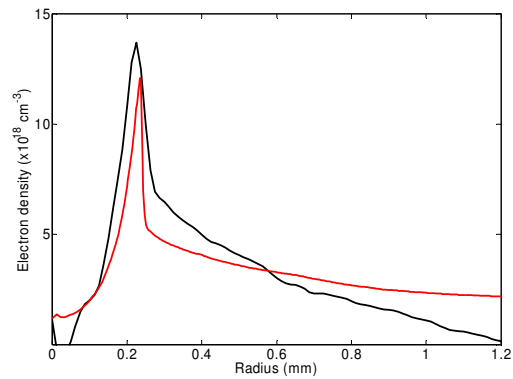


Figure 4. Electron density profiles measured in cluster blast waves at 17ns with a simulation using the NYM code (red).

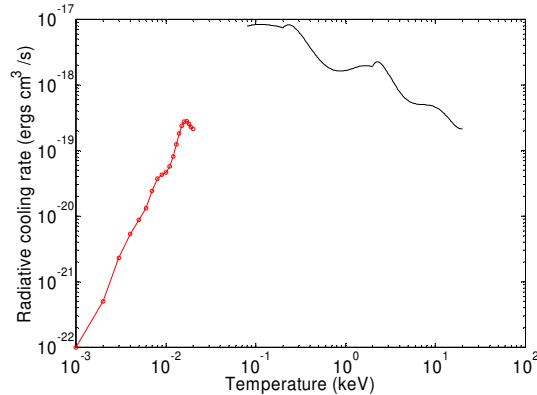


Figure 5. Radiative cooling rate for xenon calculated using RAPCAL (1 – 20eV) and the cooling rates from Ref. 20 (80 – 2000eV). The radiative power loss (ergs $\text{cm}^{-3}\text{s}^{-1}$) is obtained by multiplying the cooling rate by $\langle Z \rangle n_i^2$.

With this caveat in mind, we draw some conclusions from the values in Table 1. Immediately clear is that the radiative emission is completely dominated by line emission (bound-bound). Therefore, any use of the bremsstrahlung cooling rates used by Ryutov *et al.*^[11] or a blackbody treatment of radiative flux^[19] is wholly inappropriate. It also highlights the need for temporally and spatially resolved line emission measurements in future experiments. The radiative cooling time is extremely short so fulfils the condition of being shorter than convective timescales. However, the photon mean free path using the Planck mean opacity is $\sim 100\mu\text{m}$, similar to the scale of the shell

thickness and the precursor length, so the plasma cannot clearly be identified as either optically thin or thick. We also compare (per unit length) the radiated power loss to the kinetic energy entering the blast wave per unit time^[10] and find values of $3 \times 10^9 \text{ Jm}^{-1}\text{s}^{-1}$ and $2 \times 10^8 \text{ Jm}^{-1}\text{s}^{-1}$ respectively. This agrees with our experimental conclusion that the blast waves are fully radiative. In fact, the radiative flux is larger than the kinetic flux, possible in a blast wave because there is an initial energy provided in the explosion.

Shock instabilities

One of the main motivations for launching strongly radiative blast waves in xenon clusters is to drive a collapse of the shell and so spontaneously trigger shock instabilities. The first experiments at the Lawrence Livermore National Laboratory hoped to observe the Vishniac overstability^[26,27] which causes oscillating ripples on the shock front and is thought to be the origin of complex structure in supernova remnants. In fact the shock shell collapse was prevented by electron heat conduction thickening the shell and also heating of the precursor lowering the strength of the shock. Furthermore, in a theoretical study^[29] Laming showed a threshold velocity for the Vishniac overstability in xenon to be 25 km s^{-1} , higher than that attained in the Livermore experiment. This is somewhat of a catch-22 since to drive stronger shocks requires higher laser energies but this leads to more preheat which prevents high compression. In our recent campaign using Vulcan TAW we did see shock compressions which exceeded the non-radiative limit ($c=4$)^[36] but were probably still insufficient for the shock to be unstable to the Vishniac mechanism. The solution may be to lower the density of the target so that the gas is less opaque to the radiation. We already have evidence^[17] that this leads to higher energy loss and higher shell compression while reducing the size of the precursor.

Another instability associated with thin shell blast waves is the thermal cooling instability (TCI)^[28]. This arises when a shock enters a cycle of stall and recovery as it repeatedly loses energy through radiative losses but then rebuilds as it sweeps up the hot preshock gas. In astrophysical shocks unstable to the TCI it is predicted that the shock velocity will oscillate as it decays with a timescale of order 1000 years. To recreate this situation in the laboratory we must have energy exchange between the shocked material and its precursor. So the higher density cluster blast waves with very prominent precursors are ideal and the observation of an energy recovery phase indicates that the first stage of the process occurs even when we use low energy drive lasers ($\sim 100 \text{ mJ}$)^[9]. With the high drive energies used in the Vulcan TAW experiment, we measured full trajectories on a single shot^[16] and observed oscillations which we attribute to

this mechanism. For the TCI to occur, the cooling rate of the gas must be falling as the temperature increases (the cooling function $\Lambda(T) \propto T^\beta$ has an exponent $\beta < 1$). The radiative cooling rate for xenon is plotted against temperature in Fig. 5. The values up to 20 eV are from a RAPCAL calculation and those above 80 eV were obtained using the method of Post *et al.*^[20]. For astrophysical conditions, β is commonly less than 1 but in the laboratory this is more difficult to achieve. This calculation suggests that if we could drive xenon blast waves to temperatures above 17 eV then $\beta < 1$. The observation of the velocity instability on Vulcan was in krypton and these cooling rates are currently being calculated.

Conclusions

Since the potential of clustered gases for laboratory astrophysics experiments was recognized^[8], we have explored avenues to exploit the versatility of the method. We have demonstrated techniques which are relatively simple to perform in clusters for single-shot trajectory measurements^[16,36], creating shock collisions^[12,15], imprinting periodic shock front modulations^[15,18] and controlled pre-ionization of the upstream gas^[7]. Spatial shock front instabilities are difficult to initiate in cluster blast waves because of low compression^[9] but we have measured a velocity oscillation in krypton blast waves launched with high laser energy^[36]. Unlike the Vishniac overstability, the velocity instability cannot be studied using the stationary shock piston approach as it involves a decaying shock.

We have made progress in understanding the plasma conditions in the cluster blast waves. The system is complicated because it is not in equilibrium and cannot easily be classified as either optically thin or thick. We therefore have employed the RAPCAL atomic physics code which does not rely on these approximations. Ideally this will be coupled to simulations with non-LTE, multi-dimensional radiation-hydrocodes but this presents a difficult challenge.

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		NLTE	LTE
$\langle Z \rangle$		6.627	6.734
Fractional population of most abundant ions	Xe VII	0.356	0.312
	Xe VIII	0.518	0.529
	Xe IX	0.079	0.121
Rosseland mean opacity (cm^2/g)		1.987e3	
Planck mean opacity (cm^2/g)		1.27e5	
Radiative power loss ($\text{erg cm}^{-3}\text{s}^{-1}$)	Bound-bound	5.61e17	8.86e17
	Bound-free	4.32e13	-
	Free-free	1.20e14	-
	Total	5.61e17	8.86e17

Table 1. Calculations of radiative properties using the RAPCAL code

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Radiation Pressure effects on ion Acceleration on the GEMINI laser

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Introduction:

The dominant mechanism in most laser-driven proton acceleration experiments so far, the so-called Target Normal Sheath Acceleration (TNSA) [1], relies on the laser energy first being converted into a beam of relativistic electrons on a target foil. When these electrons exit the rear of the foil, a sheath field in excess of 10^{12} V/m is formed, which accelerates surface ions. Despite some advantageous properties [2], several factors limit the prospects of TNSA-driven beams for future applications: a relatively slow energy scaling, a large energy spread, and a low laser-to-ion energy conversion efficiency into the spectral region near the maximum energy.

Radiation Pressure Acceleration is a separate mechanism [3-6] where a large accelerating field arises by charge separation sustained by the ponderomotive pressure of intense laser radiation. Radiation pressure effects have previously been recognised as important to the dynamics of the interaction of the laser pulse with a solid target [7], leading to rapid deformation of the plasma surface and ions being driven into the target via the *hole-boring* process. In the limit of sufficiently thin foils (i.e. where the hole-boring extends to the rear target surface during the laser pulse) the laser accelerates all the ions in the laser path and the acceleration scales as if the foil were being pushed as a whole, in the so-called 'light-sail' regime. Momentum balance considerations show that the change in the momentum p of the entire foil must balance the laser momentum resulting in

$$\frac{dp}{dt} \propto \frac{2I}{\sigma}$$

i.e. the acceleration becomes more effective as the areal mass density σ is reduced, as long as the target remains opaque to the laser.

The ion energy in the light-sail regime thus scales as the square of the laser intensity - or somewhat more slowly once relativistic effects have been taken into account [8]. This rapid scaling suggests that in ultraintense interaction regimes RPA should become the dominant effect on ion dynamics in laser interactions with thin solid foils. At present intensities ($I < 10^{21}$ W/cm²), radiation pressure effects are difficult to detect. Linearly polarized pulses, normally used in interaction

experiments, lead to strong electron heating via the oscillating component of the laser's ponderomotive force [8], and to a dominance of TNSA acceleration, which masks RPA effects. The use of circularly polarized light has been suggested as a way to isolate radiation pressure effects at present intensities [4,9]. In this case, the magnitude of the laser fields is constant during an optical cycle leading to a non-oscillating, quasi-static ponderomotive push on the target electrons. A large space-charge field arises from the electrons' separation from their parent ions, while electron heating is strongly reduced [9]. However, even with circular polarization, TNSA can arise at the edge of the laser focus as the target is deformed under the spatially non-uniform pressure drive associated to typical Gaussian or Airy-type focal spot profiles. This is due to the appearance of a small oscillating electric field component normal to the surface which can accelerate relativistic electrons and drive an (albeit weaker) TNSA foil expansion [10]. For this reason, 2D PIC simulations suggest that, in realistic irradiation conditions, transition into the light-sail regime requires a stronger drive ($I > 10^{20}$ W/cm²) [4]. A further point to be taken into account is that, due to surface contaminants, most real targets consist of at least two species, which affects the acceleration dynamics [4,6,11].

We report on an experiment performed by the LIBRA consortium [12] on the ASTRA-GEMINI facility, in which we aimed to detect radiation pressure effects on ion acceleration from ultrathin foils by using ultraintense, high contrast, circularly polarized laser pulses.

Experimental Methods:

The ASTRA GEMINI laser is a Ti:Sapphire based laser which delivered up to 15J in 40-50fs during our experiment. The intensity contrast nanoseconds before the peak of the pulse was enhanced by a factor of $\sim 10^4$ using a double plasma mirror system [13] with 50-60% reflectivity. This resulted in a peak/prepulse contrast of $> 10^{12}:1$ at ns timescales and of $10^8:1$ at 4ps from the peak of the pulse. The laser was focused onto the targets at normal incidence using an f/2 off-axis parabolic mirror resulting in a $2.5\mu\text{m}$ FWHM focal spot containing approximately 40% of the laser energy in the

FWHM. Resultant peak intensities were in the range $1\text{--}5 \times 10^{20} \text{ Wcm}^{-2}$. The targets were amorphous carbon foils with thicknesses of 10-100nm. These foils have a hydrocarbon contamination a few monolayers in thickness, which is the origin of the protons observed in our spectra. The thicknesses of the foils are as quoted by the manufacturer and do not include an estimate for the hydrocarbon contamination layer that rapidly forms in all conditions other than ultra-high vacuum. A typical estimate of the contamination layer thickness is on the nm scale. The primary diagnostic for the recording of ion spectra was a Thomson parabola spectrometer, which employs parallel electric and magnetic fields to achieve energy dispersion and species separation ($B=0.8\text{T}$, $E_{\text{max}}=10\text{kVcm}^{-1}$). The particles were detected using a calibrated microchannel plate detector (MCP) [14]. Circular polarisation was used for all shots discussed in this report in order to suppress electron heating and TNSA. In order to aid the interpretation of the data, 2D Particle-in-Cell simulations were carried out using the OSIRIS code [15]. A simulation box of 20×30 microns was used with a cell size of 2.5nm . The target is a 25nm foil with a mass density of 1gcm^{-3} . In terms of composition, it consists of carbon at a density of $50n_{\text{crit}}$ and a homogeneous proton contaminant population at $10 n_{\text{crit}}$. A circularly polarized laser pulse is normally incident on the foil from the left. The pulse has a Gaussian temporal and transverse spatial profile with a spot size of $2.5\mu\text{m}$ and FWHM of 32fs . The amplitude of the pulse, in terms of normalized vector potential a_0 was 10 in the simulations of figure 2 ($a_0=eA/m_e c$, where A is the vector potential of the laser field, e and m_e are charge and mass of the electron. In practical units, $a_0=(7.3 \times 10^{-19} I \lambda^2)^{0.5}$, where the laser intensity I and the wavelength λ are expressed in Wcm^{-2} and μm , respectively).

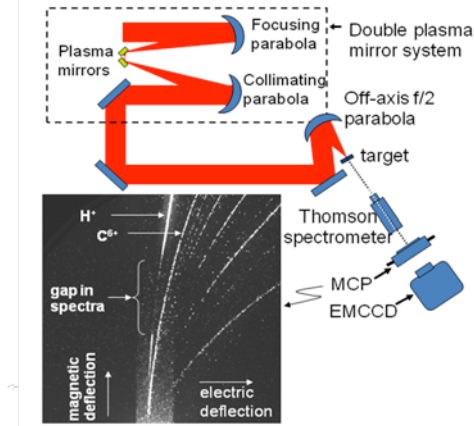


Figure 1: Experimental layout and sample of raw data. The plasma mirrors are implemented separately from the ion acceleration experiment). The raw data shows a distinct high-energy proton peak. The dominant H^+ and C^{6+} spectral tracks are indicated by arrows.

Results and Discussion:

Figures 1 and 2 show representative raw data and spectra taken at peak intensity with circular polarization. The proton spectra show peaks (~ 5

MeV in 2(a) and 9 MeV in 2(b)). In 2(b) the spectrum extends to ~ 20 MeV, and also displays a separate low energy component, with exponentially decreasing numbers up to a ~ 4 MeV cut-off. Comparison with 2D PIC simulations (figure 2 c) shows substantial agreement with the experimentally measured spectra and allows identifying the different spectral components. The low energy component is associated with TNSA-type acceleration taking place at large radii driven by hot-electrons generated at the outer edges of the focal spot (at a distance of several times the FWHM radius from its centre), while the peaked component of the spectrum is due to RPA at the centre of the spot.

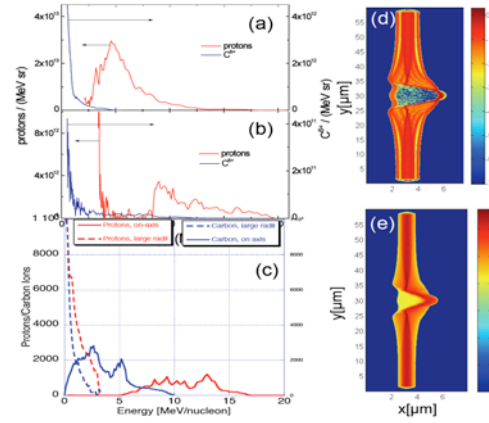


Figure 2: Comparison of experimental data and PIC simulations. **a,b** The spectra shown were recorded with circularly polarized pulses normally incident on 25nm carbon foils. Both profiles show a clear peak in the proton spectrum (red line). Such peaks were only clearly observed for circular polarisation. The inferred intensities are $1.6 \times 10^{20} \text{ Wcm}^{-2}$ and $2 \times 10^{20} \text{ Wcm}^{-2}$ respectively. **c** Simulated spectra at the end of the laser pulse for carbon ions and protons for an intensity of $2 \times 10^{20} \text{ Wcm}^{-2}$. The contributions from the central part of the laser spot only (RPA component, solid line) and the region outside the focal spot (TNSA component, dashed line) are shown separately. The combined carbon and proton spectra fit well with the observed spectra in **b**. The corresponding proton and carbon 2D density plots are shown in **d,e**. The laser is incident from the left, centered at $y=10\mu\text{m}$ and the initial foil position is at $x=3\mu\text{m}$. Comparison of **d** and **e** shows that, on axis, the protons travel ahead of the Carbon ions. The colour bars are in units of $\log(n/n_c)$, where n is the electron density and n_c is the critical density for the incident radiation.

These distinct regions of foil acceleration are identifiable in the simulated 2D density plots shown in figure 2d and e. The foil expansion far from the laser focal region is quite uniform both in the forward and backward directions and contains the full range of energies from 0 to the local maximum energy. In the central region, where the laser pressure is high enough to drive RPA there is a clear gap between the initial foil position and the accelerated foil where no protons/ions are present, corresponding to a peaked spectrum. No distinct RPA peak is visible in the carbon spectra due to a partial overlap with the low energy TNSA feature. However, close comparison of figure 2b and c indicates that the shoulder in the carbon spectrum extending to higher energies originates from RPA at

the focal spot centre, while the rapid drop at energies below 2 MeV/u is due to TNSA at the outer edges of the spot.

The spectral features observed are consistent with the detailed two-species dynamics of RPA acceleration in the light-sail regime as observed in PIC simulations [2, 4, 20]. While the force on the electrons is constant, the ions are repeatedly accelerated in the quasi-static potential gradient (effectively a self-organized double layer [27]) set-up by the laser driven separation between electrons and ions. The higher charge-to-mass ratio of protons results in the protons gaining slightly higher velocity than the C ions in the moving electric field spike, and propagating ahead of the RPA-driven Carbons. Simulations indicate that the minimum proton velocity should typically correspond to the maximum C⁶⁺ velocity [2, 4, 20], a distinctive feature of the ion spectra obtained in our experiment. By contrast, the dynamics in the *hole-boring* regime are very different [28] and should result in almost identical energies for C⁶⁺ and protons. The spectral peak features were not observed when decreasing significantly the intensity or increasing the target thickness above 25 nm, conditions which resulted in lower proton energies and continuous spectra. In all these cases, the laser intensity was not sufficient to enter the light-sail regime, and the spectral features were determined by TNSA (thicker foils typically require higher intensities in order to achieve light sail [16]. The maximum energies observed in our experiments with circular polarisation at normal incidence are ~ 140MeV Carbon ions and ~ 20MeV protons respectively.

Conclusions

Employing circularly polarized, ultra-short (50 fs), high-contrast, high-intensity pulses, we have observed novel features in the ion spectra, namely a narrow band peak in the proton spectrum correlating with the end energy of the corresponding Carbon spectrum. The scaling of the peaks (not discussed here), their dependence on polarization and target thickness, and comparison with PIC simulation results, suggest that the spectral feature correspond

to the onset of *light sail* Radiation pressure acceleration.

Acknowledgements:

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Ion acceleration from foil targets in the ultraintense ultrahigh contrast regime.

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Introduction

The acceleration of ions from the interaction of intense laser pulses with thin solid targets has generated a great deal of experimental and theoretical interest since the first multi-MeV energy ion beams were first produced in 2000 [1-6]. Those beams have shown highly desirable properties such as high brightness and low transverse and longitudinal emittance [7] with potential applications in inertial confinement fusion [8], nuclear physics [9] and medicine [10-11]. The main mechanism responsible for the acceleration of ions in laser conditions that are available today is the so called Target Normal Sheath Acceleration (TNSA), where ions from the surfaces of the target are accelerated by an electrostatic sheath field of a few TV/m. This sheath field is generated by the hot electron population, resulting from the coupling of the laser energy into the target. These electrons propagate from the front surface of the target and exit at the rear surface where they set up a large electrostatic field caused by the charge separation between the ions on the rear surface and the electrons attempting to escape from the target. The strength of the sheath field is such that the majority of the electrons are pulled back into the target. In this scheme ions with a high charge to mass ratio are preferentially accelerated, namely protons. The TNSA mechanism is the dominant ion acceleration mechanism for laser intensities ranging from 10^{18} to 10^{20} Wcm⁻² and for targets thicker than 50nm [12]. For thinner targets and higher laser intensities, new acceleration mechanisms prevail such as Coulomb explosion [13] or radiation pressure acceleration (RPA) [14-15]. Such schemes are predicted to give rise to peaked energy spectrum for the ions, with a maximum energy in the hundreds of MeV.

In this report we will report on recent experimental results obtained with the Astra-Gemini laser during the LIBRA campaign in march-July 2009 during which the acceleration of carbon ion by the TNSA mechanism has been investigated, especially the effect of the laser incidence angle. Experimental results are discussed with reference to 1D PIC simulations.

The experiment

The Astra-Gemini laser delivers pulses with duration, τ_L , of 50fs, energy, E_L , up to 12J at a wavelength, λ_L , of 800nm. A double plasma mirror system was used to enhance the contrast up to $\sim 10^{10}$ on the ps scale. The targets were irradiated under two different incident angles, 35° from target normal and 0° to the target normal. The laser pulses were focused using an $f/2$ OAP onto 1.25 μ m radius focal spot in the 0° configuration with 35% of the energy in the central spot. The calculated intensity on target was 7×10^{20} Wcm⁻².

A wide range of target materials were used during the experiment including plain foils of C, C₁₀H₁₀O₄ (CHO, mylar),

C₃H₆ (CH, polypropylene), Au and Al as well as layered Au-CH targets. The target thickness, L , was varied between 50nm and 10 μ m.

The ion beam properties were measured using two Thomson parabola spectrometers, positioned along the target normal direction for the two incidence angles as shown in figure 1.

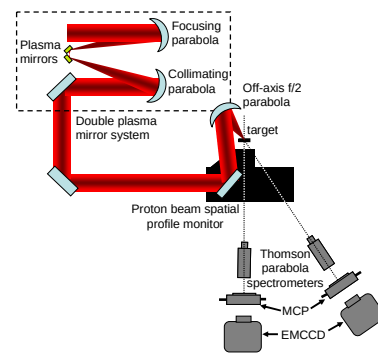


Figure 1: Schematic of the experimental setup.

A combination of Micro Channel Plate detectors and intensified Andor CCD cameras were used to record the dispersed ions. The arrangement was absolutely calibrated using slotted CR39 track detector [16].

Effect of laser incident angle on ion acceleration

Our attention was primarily focused on the acceleration of carbon ions for different target materials and thicknesses at two fixed incident angles. As shown in figure 2, clear changes in the spectra are visible when the laser incident angle changes from 35° to 0°. The number of accelerated ions as well as the maximum energy increases when the angle of incidence is changed from oblique to normal.

As shown in figure 4, this increase in maximum ion energy is independent of target thickness as well as target material. An increase by a factor ~ 1.3 is observed throughout the whole thickness range, over 2 orders of magnitude.

This result is in stark contrast with previous work by Ceccotti et al [17] where the effect of laser polarisation, s or p, on the acceleration of protons is investigated. They reported that for laser pulses of 65fs with intensities of 5×10^{18} Wcm⁻² and a contrast of 10^{10} the higher proton energy is achieved using p-polarised pulses at oblique incidence where the Brunel absorption (vacuum heating) effect [18] and the collisionless absorption mechanism [19] efficiently transfer laser energy into the target. In the case of an s-polarised laser pulse, because the laser electric field is in the plane of the target, the

mentioned absorption mechanisms have their efficiency drastically reduced, therefore diminishing the maximum proton energy.

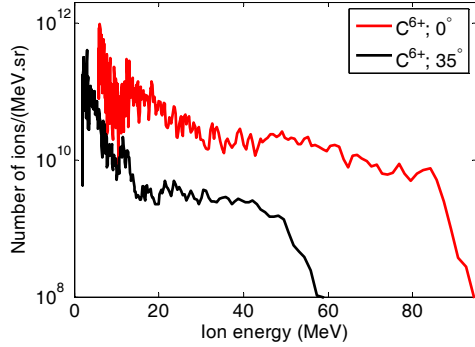


Figure 2: Experimental C^{6+} spectra obtained for two different incidence angle, 0° (in red) and 35° (in black).

In our case the laser pulse polarisation was kept constant (p-polarised) and only the incidence angle was changed. The p-polarised pulse at normal incidence is equivalent to an s-polarised pulse at normal incidence, making a comparison with Ceccotti *et al* work possible. The fact that, at higher intensities, we observe a more efficient coupling of the laser energy into the ions, which cannot be explained by the change in the focal spot size, lead us to believe that a new angle dependent laser absorption mechanism is at play.

To have a better understanding of the absorption mechanism involved in our experiment we conducted a series of 1D and 2D PIC simulations for both incident angles.

The 1D simulations were done using a modified version of the code used in [20]. A box of 25000 cells each with a size of 2nm and containing 200 particles was simulated. The target consists of two layers, one heavier ion substrate with a Z equals to 1 and a mass three times that of a proton, and a thin proton layer on the rear of the target. Both layers have a density of 90 nc. A laser pulse duration of 50fs at a wavelength of 800nm and an a_0 of 18 (equivalent to $7 \times 10^{20} \text{ W cm}^{-2}$) was used.

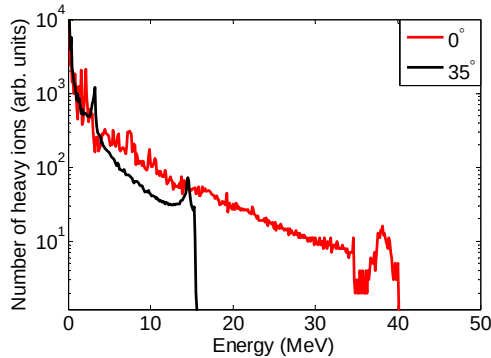


Figure 3: 1D PIC simulation result showing the energy distribution for the heavy ion population at two different incident angles, 0° (in red) and 35° (in black).

The 1D simulation results, displayed in figure 3, show a clear increase in maximum ion energy when the laser incident angle is changed from 35° to 0° . The increase in energy for the 1D case is found to be ~ 2.6 , which is slightly higher than the increase measured in the experimental data.

Effects of target properties on ion acceleration

The effect of the target properties, thickness and material, was also investigated. Figure 4 shows a summary of the measurements obtained using Al, C and CHO foils. Each point represents the average maximum C^{6+} ion energy for 3 to 4 laser shots on each target at the different thicknesses.

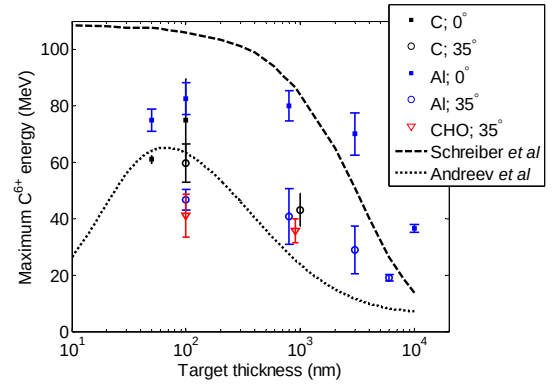


Figure 4: Average of the maximum C^{6+} ion energy over multiple shots as a function of target thickness for different materials. The errors bars represent the standard deviation of the maximum energy.

A scaling of the maximum ion energy with target thickness, and independent from target material, is observed. The increasing rate of the maximum energy is very similar to the one reported in Neely *et al* [21] for protons. An increase of a factor 2 in the maximum ion energy is observed when the target thickness is reduced by two orders of magnitude. This can be explained by the fact that the transverse spreading of the electron beam propagating through the target is increased as the target thickness increases and the effects of refluxing electrons are reduced [22], therefore limiting the acceleration of the ion by the TNSA mechanism.

Two analytical models were applied to the data in order to investigate the expected scaling of the maximum ion energy; the model presented by Andreev *et al* [23] and the model proposed by Schreiber *et al* [24]. Both models predict an increase in maximum energy with decreasing target thickness down to 100nm, in qualitative agreement with the experimental data. Andreev *et al* model predicts an optimal thickness around 80nm for our experimental parameters. On the other hand, the Schreiber *et al* model shows saturation of the maximum energy for thicknesses of ~ 100 nm. Experimentally, we observe changes in the energy spectra for targets below 100 nm, suggesting that the ion acceleration mechanism is not purely TNSA.

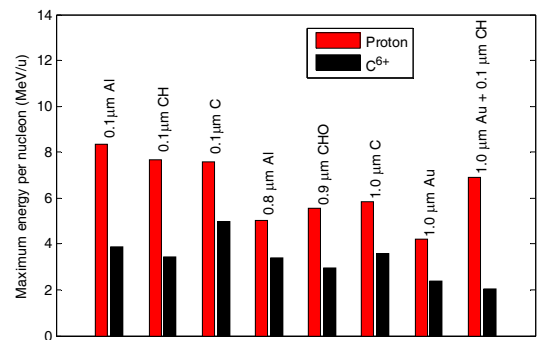


Figure 5: Maximum protons and C^{6+} ion energies for different target composition and thicknesses.

Figure 5 displays the average maximum proton and C^{6+} ion energy for different target materials at different thicknesses. This allows us to compare the effect of the presence of carbon in the target as a bulk material (C, CHO and CH targets), an uncontrolled surface contaminant layer (Al and Au) or a controlled deposited layer (Au-CH).

In the case of the C, CH and CHO targets we observed that the composition of the target does not affect the maximum proton energy significantly for a given thickness. However, in the case of C^{6+} the acceleration is more efficient for the pure C target.

This can be explained by the screening effect due to the presence of hydrogen in the CH and CHO targets. As for the controlled layer target (Au-CH), the addition of hydrogen in the accelerating field enhances the production of protons. A small decrease in the carbon ion energy is observed, further confirming that the presence of protons limits the acceleration of heavier ions.

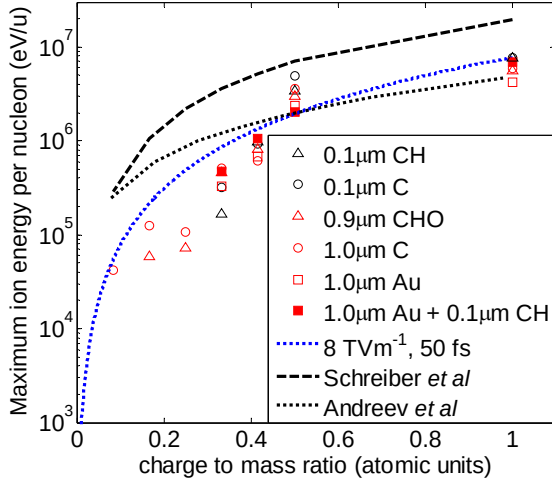


Figure 6: Maximum ion energy as a function of the ion charge to mass ratio

Figure 6 shows the variation of the maximum ion energy as a function of the charge to mass ratio. A clear increase, by a factor of 2, is observed over the range $q/m=0.1-0.5$. The analytical models by Schreiber *et al* (dashed black) and Andreev *et al* (dotted black) are also represented. The Schreiber *et al* model is in good agreement with the experimental trend but overestimates the measured energies, while the Andreev *et al* model gives energies closest to the highest measured but does not follow the trend as well. Both models fail to reproduce the lower charge state energies; shielding effects that are not taken into accounts in the models may be responsible for this. A simple model (dotted blue) where the ions are assumed to be accelerated by a simple electrostatic field and the free parameter is the electric field strength, the estimated energies for 8TV/m field is shown. This simple model reproduces the experimental data surprisingly well.

Conclusions

We have conducted an experiment using the Astra-Gemini laser in 2009 during which we investigated the effect of the laser incident angle on the ion acceleration in the TNSA regime. We observed for the first time that at the laser intensity available, $7 \times 10^{20} \text{ Wcm}^{-2}$, the coupling of the laser energy into the ions is greater at normal incidence than at oblique incidence. This observation is inconsistent with previous work by Ceccotti *et al* [17] intensity. The increase in maximum ion energy can not be explained by the small difference in laser intensity at the two angles, therefore suggesting that a new angular dependent absorption process may occur at these higher laser contrasts and intensities.

1D PIC simulations were performed to investigate further this possible new absorption mechanism. The experimental data were well reproduced by the simulations. 2D simulations were also conducted to explore the effect of the incident angle on the electron population. Preliminary analysis suggests that higher energy fast electrons are produced in the normal incidence case.

We also found that with the laser parameters available during the experiment the maximum ion energy increases with decreasing target thickness down to 100nm independently from target material for the TNSA mechanism. For thinner targets,

changes in the energy spectra were observed suggesting that the acceleration mechanism is not purely TNSA.

The investigation of the effects of the target material on the acceleration of carbon ions showed that the highest energy carbons are obtained with targets containing carbon but no hydrogen as a bulk material. The presence of hydrogen, throughout the target or on the surface reduces the energy coupling to the carbon ions. By contrast, it is found that the acceleration of protons can be enhanced for a high Z material when a hydrogen rich layer is deposited on the back surface.

An increase in the maximum ion energy with increasing q/m ratio was observed and analytical models were applied to the data. It has been found that a simple electrostatic acceleration model gives a reasonable fit to the data as a function of q/m . Unconsidered screening effects, in the models, of the lower ion charge states may explain the difference between the models and the data for these ions. For further reading please see [25].

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Characterisation of debris emission from thick targets on Astra-Gemini

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Introduction

The issue of debris from solid targets has been a concern since the advent of higher power lasers where protective barriers, such as glass pellicles [1], are used to protect optics. However with high repetition rate high power laser systems now coming online or in the design stages [2-4] the issue of debris is becoming more significant [5] as the accumulation of damage or surface coating is more rapid.

We report we report on debris characterisation measurements made during the radiation commissioning of the Astra-Gemini laser in early 2009. During the commissioning the distribution of debris from thick targets around the horizontal plane of the target was studied. Debris in this report is defined as to encompass all material leaving the target; liquid, vapour and solid (shrapnel).

Experimental setup

The Astra-Gemini laser delivers pulses with duration, τ_L , of 50fs, energy, E_L , up to 15J at a wavelength, λ_L , of 800nm. The laser incident angle onto target was 35° relative to the target normal. The laser pulses were focused using an $f/2$ OAP onto 1.25 μm radius focal with 35% of the energy in the central spot. The calculated peak intensity on target is $6 \times 10^{21} \text{ Wcm}^{-2}$.

The targets used are 0.5 mm thick Tantalum (Ta), plastic (CH) and copper (Cu) and 50 μm thick gold (Au). The key diagnostics are four 127 mm square glass witness plates [ref RAL report] arrayed around the target as shown in figure 1. These glass plates accumulate debris on their front surfaces over shot sequences consisting of up to 30 shots.

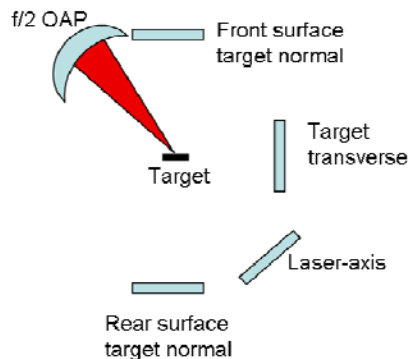


Figure 1: Schematic of the experimental setup.

These witness plates enable the study of three different debris types on surfaces. These are the build up of layers of debris coating on surfaces, macro sized pieces of debris embedded on surfaces and the damage caused by shrapnel like debris to the surfaces.

Table 1: Witness plate positions and distances, d , from target

Witness plate position	d (mm)	Solid angle (sr)
Front surface target normal axis	320	0.16
Target transverse	243	0.27
Laser axis target rear surface	230	0.30
Rear surface target normal	245	0.27

Debris from 500 μm thick targets

The debris generated from 500 μm thick targets of Ta, CH and Cu were accumulated on a single set of witness plates. There were in total 36 shots on CH, 48 shots on Cu and 47 shots on Ta. As these shots were primarily for the radiation commissioning there are shots within this total that were defocused, table 2 summarises this for each target, though the majority were at best focus.

Table 2: Number of shots at different focus positions for the three materials used for 500 μm thick targets

Material	Estimated spot diameter (μm)							
	3	10	20	30	50	100	250	500
Cu	22	4	4	0	4	4	6	4
CH	12	4	4	0	4	4	4	3
Ta	38	1	3	1	2	0	0	0

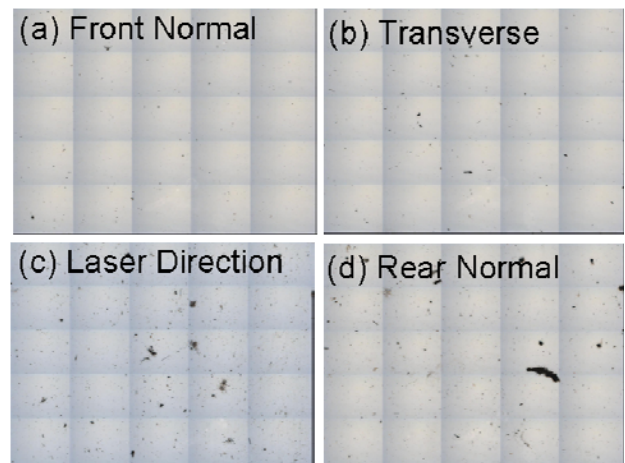


Figure 2: Scans of central area (15.9 x 11.9 mm) for each witness plate for 500 μm thick targets. (a) Front surface target normal, (b) target transverse, (c) laser axis and (d) rear surface target normal.

The debris distribution for this thickness of targets was found to favour emission in the laser-axis and rear surface target normal directions which showed the highest density of debris. This can be seen in Figure 2 which shows systematic scans of the central area for each of the witness plates, covering an area of 189 mm².

The debris particulates observed on the witness plates are large, typically around 400 µm. The debris is typically irregular in shape which is especially true for plastic. The plastic target was observed to produce the largest pieces of debris that stuck to the witness plate with sizes in the millimetre range (figure 2(d) shows one of these large pieces). For the metal targets if any such large pieces of debris were produced they were not captured by the witness plates. No large scale physical damage to the witness plates surfaces were observed

Debris from 50 µm thick targets

A second set of witness plates were used to capture the debris produced from the 50 µm Au targets shot during the radiation commissioning. The positions of the witness plates were kept the same as shown in figure 1 and listed in table 1. As before, multiple shots worth of debris were accumulated onto the witness plates, in this case the number of shots was 26. Within this were shots taken at different focus positions and are listed in table 3.

Table 3: Number of shots at different focus positions for the 50 µm thick Au targets

Material	Estimated spot diameter (µm)							
	best	10	20	30	50	100	250	500
Au	2	4	4	0	4	4	4	4

The distribution of debris from the thick 50 µm Au targets is found to be different to the case for the very thick 500 µm targets. For the thinner target the highest debris was found on the front and rear target normal witness plates and minimal amount on the laser axis and transverse plates. The largest debris particulates measured on the laser axis and transverse plates is 70 µm and 20 µm respectively. Scans of the central area for each of the witness plates are shown in figure 3.

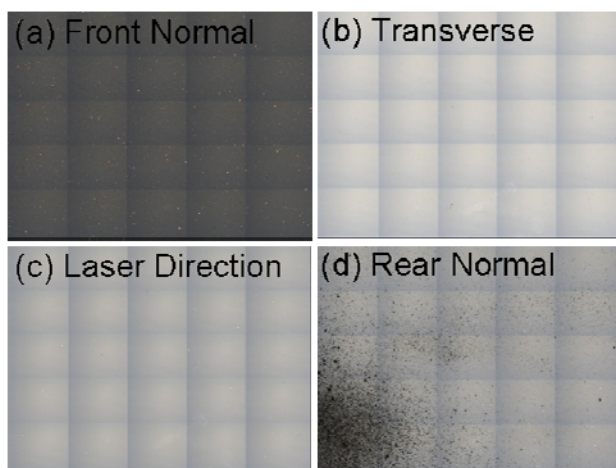


Figure 3: Scans of central area (15.9 x 11.9 mm) for each witness plate for 50 µm thick Au targets. (a) Front surface target normal (note this image is reflected rather than transmitted light to highlight gold debris), (b) target transverse, (c) laser axis and (d) rear surface target normal.

The debris on the front surface plate was found to be evenly distributed across the surface. The size of the debris particulates was found to be smaller in size than for the larger targets, largest debris size was found to be ~70 µm in size. The debris

was also found to be more uniform in shape, circular and flat on the glass surface. This is likely due to the Au material hitting the plate being molten droplets rather than solid shrapnel. The rear surface target normal plates

The rear surface target normal plate has a similar layer of evenly distributed debris across the surface as the front surface but also has an additional feature. This additional feature is an area of damage to the glass surface in a concentrated region around the actual target normal axis. This can be seen in the bottom left of figure 3(d). This indicates a beam like feature of highly destructive debris, shrapnel, coming from the target rear surface. This area of damage has distinct features, a central area where the pits in the glass are ~350 µm in size, two larger circular areas on damage above and below the central area which contain pits smaller in size of ~100 µm. The final feature is a line of damage from the central region going towards the laser axis with pits also around 100 µm in size. These features are shown in figure 4.

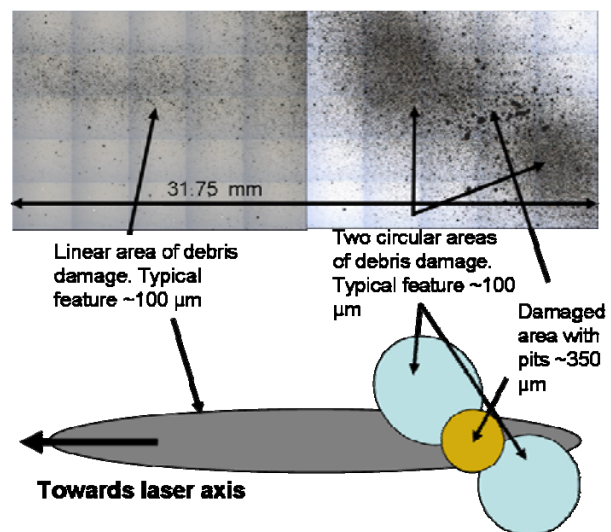


Figure 4: Debris damage to glass witness plate at the target normal rear surface position. The images at the top are scans of the damage to the plate surface and the bottom diagram is a schematic of features found in the damage distribution.

Debris production with the Phelix laser

Similar debris damage along the target normal rear surface axis was also observed on an experiment using the Phelix laser, based at the GSI facility in Germany.

The Phelix laser generates pulses that are of the order of a picosecond with energies in the 100 J range. The debris deposition along the target normal axis from 26 µm and 100 µm Au targets was obtained from the front Al layer of an RCF stack (50 mm x 50 mm) placed 40 mm behind the target, which acted as a witness sample.

It was observed that for 100 µm thick Au targets indents were formed in the Al layer. It was confirmed that these indents were being formed by Au debris from the target as Au shrapnel was found to be embedded in the 13 µm thick Al layer. This can be observed in figure 5(b).

When the 26 µm thick Au target was irradiated the results were drastically different in terms of debris damage. Instead of the indentation caused by debris impact it was found that a roughly radial symmetric discolouration of the Al layer had occurred, this can be seen in figure 5(a). This discolouration is purple in colour near the centre and gradually changes to a golden yellow at larger radii. This is an indication of heat damage to the Al layer and is likely due to non-shrapnel debris. Modelling is underway to determine the reasons for the change in size of debris particulates with target thickness.

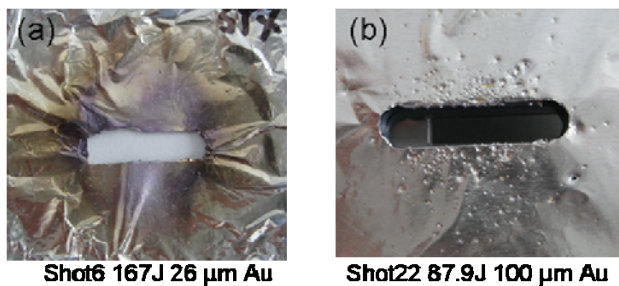


Figure 5: Representative samples of the effects of debris on an Al witness foil placed along the target normal at the rear of: (a) 26 μm Au and (b) 100 μm Au targets. The thicker target produces shrapnel.

Conclusions

We have taken debris measurements with glass witness plates on Astra-Gemini during radiation commissioning and observed the effects that target thickness has on debris.

For 0.5 mm thick targets, large debris particulates were measured over a wide area with the highest density being measured in the laser axis and rear surface target normal directions. It was found that the debris particulates were not energetic enough to cause physical damage to the glass witness plates at a distance of ~ 25 cm. It was also found that the largest pieces of debris that stuck to the glass surfaces were from plastic targets.

For targets that were an order of magnitude thinner, 50 μm Au, the distribution of debris and its effects were found to have changed. It was weighted more towards the front and rear surfaces target normal axes. The debris at the front surface was found to form a coating on the glass witness plate consisting of circular discs, indicating the debris was molten or liquid when impacting on the plate. Similar debris coating at the rear surface was also found, but in addition severe damage to the glass surface was also observed. This indicated that the debris at the rear surface consisted of a mixture of shrapnel and liquid or molten pieces. Features were found in the distribution of damage to the glass surface consisting of a central area with largest damage, two surrounding hemi-spheres of lesser damage and a line of damage stretching towards the laser axis.

Similar damaging shrapnel was also observed on the Phelix laser for 100 μm thick Au targets. When the laser energy was increased and the target thickness decreased there was significantly less physical damage observed but thermal discolouration also occurred.

These initial observations indicate that the target thickness has a significant effect on the debris distribution and types of debris produced in the interaction environment. However, a more systematic study of debris is required to determine how changing laser and target parameters effect debris distribution and types. In particular the debris produced from ultra-thin targets, i.e. submicron, should be investigated as these targets are likely to have very different debris distributions and are the favoured target type for achieving the required ion beam properties for application [refs].

Acknowledgements

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High rep rate Thomson parabola-MCP assembly for multi-MeV ion spectroscopy

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Abstract

We report on the absolute calibration of a micro channel plate (MCP) detector, installed as detector in a Thomson parabola spectrometer. The calibration delivers the relation between a registered count numbers in the CCD camera (on which the MCP phosphor screen is imaged) as a result of the impact of an ion beam onto the MCP. The particle response of the whole detection system was evaluated by using laser accelerated ions with proton energies up to 3 MeV and carbon ion energies up to 16 MeV. In order to obtain an absolute measurement of the number of ions incident on the MCP detector, slotted CR-39 track detector was installed in front of the MCP. The signal registered on the MCP due to ions propagating through the CR-39 slots is compared to the number of particles counted on the adjacent CR-39 stripes after the etching. The calibration of the response of MCP has been extended to higher energy ions and protons on the basis of a simple model validated by comparison with the calibration data. This sensitive detection set-up makes it possible to measure in a single laser shot the ion spectrum in absolute terms.

1. Introduction

High power (100TW–1PW) laser systems are capable of delivering ultraintense (up to 10^{21} W/cm²), ultrashort (40–50 fs) laser pulses at high repetition rate (e.g. Astra-GEMINI laser-one pulse in every 20s). Energetic ions/protons accelerated up to tens of MeV energies per nucleon have been observed experimentally during the interaction of these pulses with thin solid targets [1–3]. Efficient detection of these particles is most important for their potential applications. The Thomson parabola spectrometer is successfully used as a charged particle analyzer [4] for the quantitative analysis of ion beams emerging from the laser plasma. The deflection of charged particles in homogeneous magnetic and electric fields provides simultaneously their energy, momentum, and mass-to-charge ratio. The spectrometer becomes uniquely valuable, particularly for use on a single shot basis, if coupled to absolutely calibrated, fast response detectors, such as, e.g. microchannel plate (MCP) detector coupled to a phosphor screen. Having a single shot online diagnostic is very important also because

current laser systems still suffer from shot-to-shot fluctuations of laser pulse parameters, such as energy, pulse duration, and pulse shape, which significantly modify the interaction conditions and, thereby, the particle acceleration processes. This paper will describe the calibration of a Thomson-MCP assembly which we employed in a recent laser-driven ion acceleration experiment. We will discuss the method employed, the relevant data analysis and a theoretical model for the MCP response which allows to extend the calibration data to higher energy ranges. Careful consideration is required in evaluating the response of the MCP, since in the arrangement employed different energy ions are incident on the MCP at different angles (unlike, for example, ions accelerated by linear accelerators), and this may affect the gain and secondary electron yield. For this reason, an in-situ calibration is important for a correct data analysis.

2. Method and calibration

In the experiment, thin planar foil targets were irradiated at 35° incidence by p-polarized laser pulses from the Astra-GEMINI laser system at the Rutherford Appleton Laboratory (RAL) with energy of up to 12J and pulse duration ~50 fs. With an f/2 off-axis parabolic mirror, a maximum vacuum intensity of $5 \cdot 10^{20}$ W/cm² was reached with a double plasma mirror system. The measurements of the energies of the ions emitted normal to the target were carried out with a Thomson parabola spectrometer schematically shown in Fig. 1. Typically, a magnetic field of about 0.4T and electric field of 8 kV/cm have been applied. The ions are detected by a Hamamatsu MCP (Model-AF2226-A093B) detector with a diameter of 77mm and bias angle 8° coupled to a phosphor screen (P-43) which is imaged with an objective onto the chip of a CCD camera. In order to calibrate the MCP we placed a slotted CR-39 track detector [5] in front of the MCP. With this arrangement the spectral parabolic traces of the ions will be partly detected on CR-39 while the parts, which pass through the gaps of the slotted CR-39, will appear on the phosphor screen of the MCP. The tracks on CR-39 produced after etching in a 6N NaOH solution at 70° C temperature of water bath for 3 hrs and the corresponding image from the phosphor screen of the MCP detector captured on CCD camera

are shown in Fig.2(b) and (a), respectively. The number of protons and C^{6+} ions incident on each stripe on CR-39 were counted using an optical microscope where as a routine in Matlab was written to identify the ion species and their energies by fitting the different parabolic tracks from the image. This routine also provides the counts in each pixel of the CCD as a signal corresponding to the ions falling in that pixel region. We correlate the number of protons on each stripe to the adjacent CCD integrated counts belonging to the same parabolic trace. A complication encountered was that the portions of the traces projected on the CCD through the CR-39 gaps for the lower energy part of the spectrum appeared smaller than their counter part on CR-39. This was due to the thickness (1mm) of the CR-39, coupled to the relatively large deflection angle of the low energy ions, which resulted in part of the trace being blocked. Therefore we considered the counts per unit energy interval integrated across the width of the stripe on the CCD image and the corresponding integrated number of protons on CR-39. In order to correlate CCD counts

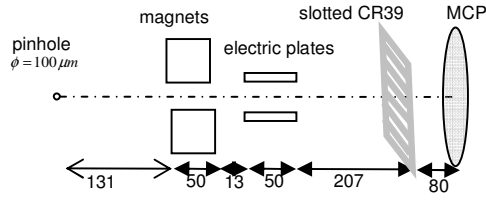


Fig.1 Schematic arrangement of the Thomson parabola spectrometer to detect the ions using the MCP detector coupled to the phosphor screen and CR-39 detector simultaneously (Distances in mm).

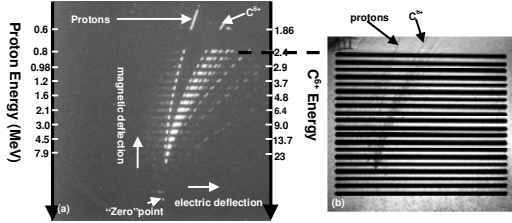


Fig.2 An image from the (a) MCP-phosphor screen and (b) CR-39 track detector of an emitted ion spectrum from 0.8 μ m carbon foil target taken from a single laser shot in the target normal direction with the Thomson spectrometer. Here the CR-39 track detector was installed in front of MCP detector.

to number of protons we then interpolated the signal of two consecutive trace sections on the CCD and compared the interpolated value with the counts of the interleaved section on the CR-39. Similarly, interpolation of two consecutive CR-39 trace sections and comparison with the interleaving CCD trace section was also carried out. In this way we obtained the ratio (counts/particle) as a function of energy which gives the response of the MCP to protons. In the same manner the response to C^{6+} ions has been calculated. Data obtained from the comparison of

spectral images on MCP and CR-39 is shown in Fig. 3 for protons and C^{6+} ions. Although the calibration data shown are for protons up to 3 MeV and for carbon ions up to 16 MeV energies, the MCP response to higher energy ions was considered and a theoretical model has been proposed.

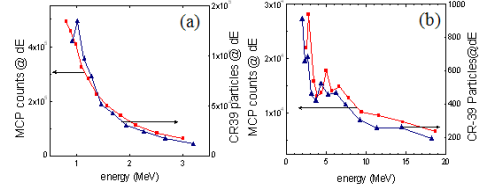


Fig.3 Correlation of integrated counts in dE energy interval due to MCP signal with number of particles on CR-39 in dE energy interval plotted with respect to energy for (a) Protons and (b) Carbons. The dE energy interval corresponds to thickness of the stripe on CCD and CR-39

3. Theoretical Model

In general the response of the MCP to the incident ion can be given as a function of channel bias angle, open area ratio, incident angle of the ions on channel, their energy and the penetration depth into the channel. All these parameters some how affect the secondary electron yield due to the ion impact and therefore the gain of the MCP channel. Following Beuhler and Friedman [6] a general expression for the secondary yield from a solid target can be given by

$$\gamma_a = \frac{P}{E_0} \int_0^\infty e^{\frac{x \cos \theta}{\lambda}} \left(\frac{dE}{dx} \right)_e dx \quad (1)$$

where P is the probability of electron escape from the surface, E_0 is the energy (eV) needed to be deposited to produce an electron for escape, λ is the characteristic length of electron diffusion inside the target, $(dE/dx)_e$ is the electronic stopping power of the projectile ion into the target and θ is the angle of incidence with respect to the normal to the target surface [7]. Several investigations have shown experimentally [8] and by numerical simulations [9] the dependence of secondary yield γ_a on the electronic stopping power of the projectile ion $(dE/dx)_e$ over a broad range of ion energies. In addition, the angular dependence of the secondary yield $\gamma_a \propto 1/\cos \theta$ was shown experimentally by Thieberger et al [10] for carbon ions. In our model calculations we have considered the same dependences. Assuming in (1) that P , E_0 and λ are constant for a particular material and $(dE/dx)_e$ can be considered constant as well for the electron escape from a very thin layer at the surface of the material, Eq.(1) becomes

$$\bar{\gamma}_a = \frac{P}{E_0} \frac{\lambda}{\cos \theta} \left(\frac{dE}{dx} \right)_e \quad (2)$$

The gain of the MCP channel is normally given as $g = e^{G/a}$, where G is the secondary emission characteristics parameter and $a = L/D$, where L is the channel length and D is the channel diameter [11].

Implicitly the gain depends on the voltage across the channel and a linear dependence of the voltage on channel length could be considered. In this way the gain has been calculated to be, $g = e^{k \cdot (L - z_{pd})/L}$, where z_{pd} is the penetration depth and k is constant. In our case, since the MCP is installed in a Thomson spectrometer, and therefore the angle of incidence of the dispersed ions on the MCP differs for different energies, the penetration depth z_{pd} is also different, and therefore the gain will be in principle different. A Monte-Carlo simulation has been therefore performed in order to obtain the most probable penetration depth and consequently the gain in the channel. We have considered the intersection of straight line ion trajectories with cylindrical channels whose direction cosines incorporate the incidence angle and channel bias angle. Finally the response of the MCP can be written as

$$\text{Response}_{\text{MCP}} \propto \frac{1}{\cos \theta} \cdot \left(\frac{dE}{dx} \right)_e \cdot g \quad (3)$$

To obtain $(dE/dx)_e$ for different energy ions we used the SRIM [12] program and the angle of incidence defined by the geometry of the experiment. For C^{6+} ions θ was $\sim 82^\circ$ at 6 MeV and $\sim 89^\circ$ at 240 MeV and for protons θ was $\sim 82^\circ$ at 0.8 MeV and 87° at 5 MeV. As discussed above, in calculating the $1/\cos \theta$ dependence, we take into account that the incidence angle θ with respect to the normal to the MCP channel surface varies for different ion energies in the spectrometer. The model provides a good agreement to our data except for the lower energy proton range (below 1.5 MeV). The experimental calibration data at higher energy range is shown in Fig. 4(a) and (b) together with the model calculation for the response of the MCP. We see in Fig. 4(b) a good agreement for carbon ions and a change in response by a factor of 1.5 passing from 20 to 240 MeV. However there is a disagreement for the lower energy protons (< 1.5 MeV) between the theory and experimental data shown in Fig. 4(a) which we are currently investigating. We note that the relative error in this part of the spectrum is higher (about 20%) due to the

higher percentage error in determining the energy range.

4. Conclusions

In conclusion, in this paper we have described the measured particle response of an MCP detector employed within a Thomson spectrometer. The calibration delivers the relation between a registered count numbers in the CCD image of the phosphor screen which results from ion impact. The particle response of the whole detection system was evaluated by using a laser accelerated ion beam containing protons up to 3 MeV and carbon ions up to 16 MeV energies. Based on a model for the MCP response the calibrations have been extended to higher energies range. The data analysis has highlighted the dependence of MCP response on incident ion species. This sensitive detection set-up makes it possible to measure on a single shot basis the ion spectrum in absolute terms.

Acknowledgements

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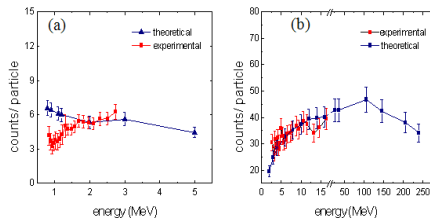


Fig.4 Response of MCP (counts/ particle) (a) for Protons and (b) for C^{6+}

Proton/ion energy scaling and laser conversion efficiency using 50fs, 10^{20} - 10^{21} W/cm² Astra-Gemini pulses

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Introduction

The laser acceleration of ions to multi-MeV energies from thin foils has been investigated extensively during the last decade using intense laser pulses (10^{18} – 10^{20} W/cm²) [1]. The ions are mainly accelerated in the space-charge fields created by laser generated relativistic electrons which are penetrating the target (e.g. Target Normal Sheath Acceleration – TNSA mechanism [2-5]). Experimental results have shown that these ions have unique properties: high brightness (about 10^{13} protons/ions per shot), high current (in kilo-Ampere range), ultralow emittance, and short pulse duration (less than 1ps), opening prospects for a broad range of applications [1]. While they have been applied successfully in high resolution radiography [6], most potential applications are demanding further improvement of the beam specifications particularly regarding maximum ion energy and ion flux.

Currently the highest proton energies (~60 MeV) and conversion efficiencies (8%) have been obtained using Nd:Glass PW systems providing 100s J, ~ ps (or several 100s fs) pulses. These systems are large-scale, costly installations, with very low repetition rate. The use of smaller scale (possibly table-top), high-rep systems is clearly preferable in view of widespread applications. For example, Ti:Sa systems reach high intensities by concentrating more moderate amounts of energies in very short pulses (10s of fs). Technological progress in this area is fast and is now enabling access to unprecedented intensities (above 10^{20} W/cm²), with ultrashort (~50 fs) laser pulses [7]. The interaction at these intensities still has to be explored carefully and experiments aiming to obtain scaling laws or estimates of the efficiency of the acceleration process are essential. The scaling of proton energy using ~ 50 fs lasers at intensities ranging from 10^{18} to 10^{19} W/cm² has been reviewed in [8, 9]. Generally, acceleration with shorter pulses is, at comparable intensities, less efficient than with ps pulses. By optimizing target thickness (down to tens of nm) [8, 10, 11], maximum conversion efficiencies into protons of about 1 % have

been inferred.

Additionally, PIC simulations using circularly polarised light and intensity $>10^{20}$ W/cm², have shown a transition from the usual TNSA process to a regime of radiation pressure acceleration (RPA) [12, 13]. The advantage of using circularly polarised pulses lies in the fact that the oscillating components of the Lorentz force in the direction perpendicular to the sharp density gradient is quenched, and hence the motion of the electrons at the interaction surface is mostly adiabatic and electron heating is strongly reduced [12]. The ions are accelerated via space charge fields which balance the radiation pressure (i.e. the ponderomotive force).

In this report we will discuss the effect of the laser polarisations, target thickness and target material on the energy of the accelerated protons/ions under normal laser incidence on the target. Also we will discuss the total number of protons/carbon ions produced during the interaction at two different laser incidence angles namely 35° and 0° for a range of target thickness.

Experimental Method

The experiments have been carried out on ASTRA-Gemini laser at Rutherford Appleton Laboratory, which delivers 12 J ultra-short (~50 fs) pulses at a central wavelength of 800 nm. The intrinsic intensity contrast of 10^7 at 20 ps prior to the pulse peak was enhanced to the level of $\sim 10^{10}$ employing a “double plasma mirror” system, which preserves the spatial focal spot qualities although the throughput laser energy is reduced to ~6J. An f/2 off axis parabola was used to focus the laser pulses to a spot size of diameter $\sim 2.5 \mu\text{m}$ containing 35% of laser energy.

Thus, intensities above 5×10^{20} W/cm² could be reached. Al and carbon targets with thickness varying from 10 nm up to $10 \mu\text{m}$ were irradiated up to this intensity. Thomson spectrometers with absolutely calibrated micro-channel-plate (MCP) detectors registered ion emission spectra simultaneously along different directions: along the laser propagation axis

(0°), and at angles of 35° , 45° and 215° relative to the laser axis. The spectrometers employed 100 μm -diameter entrance pinholes located at a distance of 130, 93, 95 and 81 cm from the target surface, respectively. The schematic of the experimental set up has been shown in fig1.

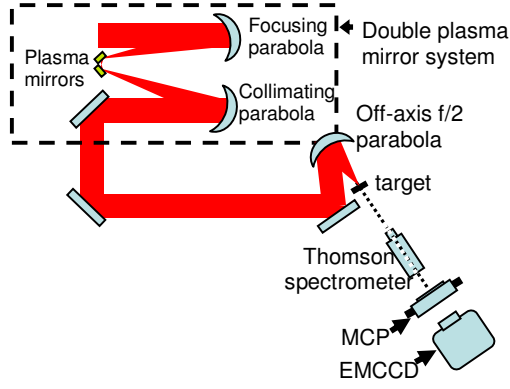


Fig.1 Experimental setup: A double plasma mirror was employed to enhance the laser pulse contrast.

Results and Discussion

The dependence of the maximum proton energy on carbon target thickness at two different polarisations viz. linear and circular at normal incidence is shown in fig 2(a).

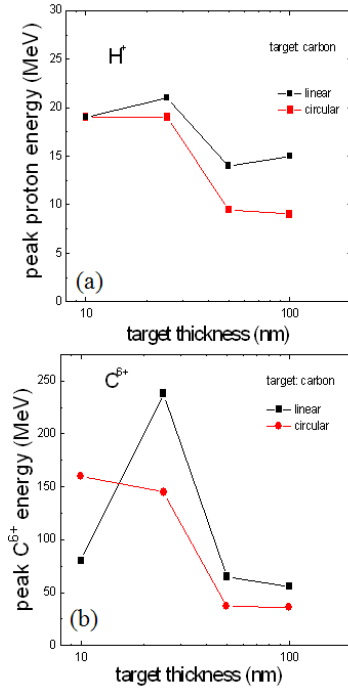


Fig.2 Maximum proton/ion energy dependence on target thickness at normal incidence on carbon target for linear and circular polarisations

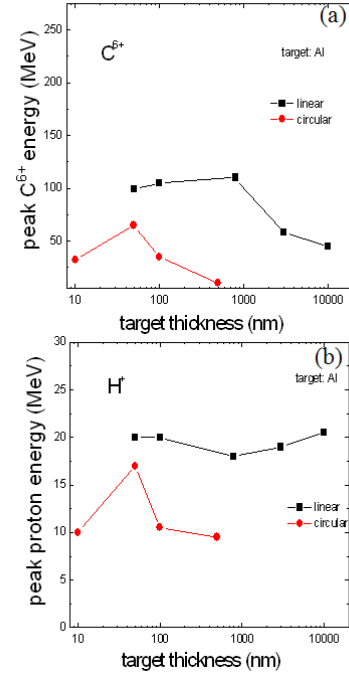


Fig.3 Maximum proton/ion energy dependence on target thickness at normal incidence on aluminium target for linear and circular polarisations

Proton energy increases with decrease in target thickness for both polarisations and there is factor of 2 gain in energy (~ 20 MeV) at 25nm compared to 50nm for the circular polarization case (~ 1.5 times gain in linear case).

Similarly in fig2(b) we have shown the carbon ions energies dependence on thickness for two polarisations. At circular polarisation, the C^{6+} energy increases with decrease in thickness and is ~ 160 MeV at 10 nm with a ~ 4 times increase in energy going from 100nm to 10nm. With linear polarisation, energy increases with a decrease in thickness up to 25 nm reaching ~ 240 MeV.

Next, we investigated the dependence of proton/ion energy on the target thickness but now with aluminium target at normal incidence. In fig3(a) the proton energy dependence on thickness at circular and linear polarisation is shown. Similar to the case of fig 2, for circularly polarized light the proton energy increases with decrease in thickness. Surprisingly for linear polarisation we see an almost constant maximum energy ~ 20 MeV at all thicknesses ranging from 10nm to 50nm. On the other hand in fig 3(b) C^{6+} energy has been plotted with target thickness. For both polarisations we see an increase in the energy as thickness decreases.

The dependence of produced number of protons/ions at two different laser incidence angles 35° and 0° on the target as a function of target thickness for the aluminium target has also been investigated. In fig.4(a) the total number of protons and carbons detected along the rear surface target normal (RSTN)

within a solid angle of 9 nsr (hereafter we will label this quantity as proton/ion flux) has been plotted against target thickness under 35° laser incidence. The proton/ion flux increases with decrease in thickness up to 100nm and decreases below this thickness. The numbers are integrated over the 2 - 12 MeV range for protons and 2.5 - 42 MeV for Carbon ions. Both, proton and ion flux increased by an order of magnitude when the target thickness was decreased about 200 times. One can try to estimate the conversion efficiency into protons for 100 nm thick targets by assuming a proton beam divergence of about 8° (as suggested by other diagnostics) which gives ~ 6.5% (~ 5% for carbon ions).

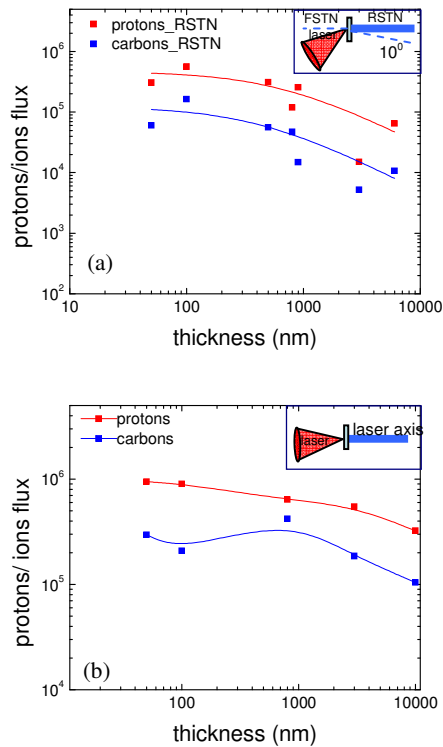


Fig.4 Number of accelerated protons and carbon ions as a function of target (Al) thickness (a) Along rear surface target normal (b) along laser axis forward direction

Conclusion

In summary we have presented the energy scaling for protons and carbon ions as a function of target thickness for two different target material (Al, C) and two different polarisations (linear and circular). Also the dependence of produced proton/ion flux was shown for two incidence angles namely 35° and 0°. The interpretation and modelling of this data is currently ongoing.

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Investigation of Contrast of Astra/Gemini

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Introduction

With remarkable progress in chirped pulse amplification (CPA) technology, extreme laser power and intensity as high as 10^{22}W/cm^2 has recently been made available by the new generation femtosecond (fs) petawatt (PW) Ti:Sapphire amplifiers [1-4]. Such ultrahigh intensities have opened a way to the experiments in the ultra-relativistic and radiation dominated regime of laser-matter interaction [5-6]. The temporal contrast of the laser pulse, defined as the ratio of the peak intensity of the pulse to the prepulse intensity, plays a crucial and important role in the ultra high field laser-matter interaction experiments. It generally requires a clean laser pulse with a contrast better than 10^{10} to restrict any destructive preplasma dynamics in those experiments, as excessive prepulse intensity can significantly modify the solid targets and experiment conditions due to preplasma formation prior to the arrival of the main pulse.

There are two major types of prepulses in the temporal profile of ultrafast, high power laser pulses: the incoherent and coherent pedestals. The first is normally caused by the amplified spontaneous emission (ASE) on the ns time scale while the second, on the order of tens of ps time scale, is related to the residual high order spectral phase, spectral phase noise, random spectral modulation and degree of non-Gaussian shape introduced through the CPA laser system. The incoherent ASE contrast is generally considered to be more detrimental than the coherent contrast as it is on the relatively long ns time scale which could result in sufficient preplasma expansion to alter the experimental conditions significantly [7-8]. To meet an increasing demand for higher temporal quality of the laser pulse by considerably reducing both the incoherent and coherent noise level has posed a big challenge to the development of extreme power laser systems.

In this article, we have report an investigation of the temporal contrast of the Astra-Gemini high power Ti:Sapphire laser. Enhanced contrast of the laser pulse by nearly an order of magnitude has been achieved by upgrading the commercial front-end to provide cleaner seed pulses.

Results and discussion

Astra-Gemini, as a world leading PW high power solid-state laser facility, has been in operation successfully for more than two years. The Astra laser is a well established high power Ti:Sapphire system based on CPA technology. This laser system consists of a commercial front-end including a Ti:Sapphire mode-locked oscillator and a 9-pass Ti:Sapphire amplifier (Femtolaser) operated at 1kHz; a grating pulse stretcher; a chain of three multi-pass Ti:Sapphire amplifiers working at a repetition rate of 10Hz and final recompression by a pair of gratings. Pulses of duration $\sim 45\text{fs}$ at energies up to 0.7J was delivered at 10Hz. An acoustic-optic programmable dispersive filter (AOPDF, Dazzler) is also used in the front end to control the overall spectral bandwidth and compensate the residual high order phase. Detailed information on

configurations, geometries and specifications of the Astra laser facility can be found elsewhere [3-4].

In order to improve the contrast of the laser pulse to meet the requirement for high field laser-matter interaction applications, we undertook an extensive study of temporal profile of the Astra laser pulse. Initially, the temporal profile of the pulse from the existing system was measured with a commercial third-order auto-correlator (SEQUOIA) to provide a baseline of comparison for further optimisation and improvement. A typical contrast measurement of the Astra laser pulse is shown in Figure 1.

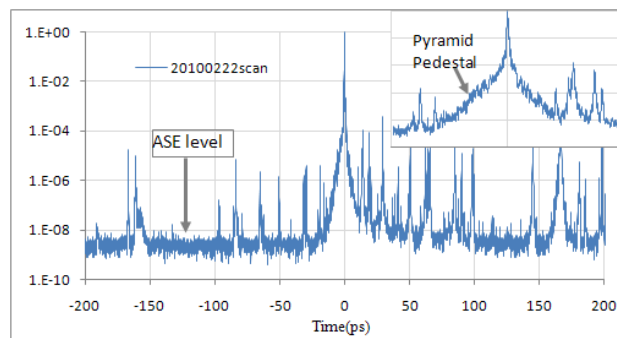


Fig.1 Typical temporal profile of the Astra pulse

As seen, the background noise level of the laser pulse on the ns timescale is in the region of $\sim 3 \times 10^{-9}$ compared with the main peak, corresponding to an incoherent ASE contrast of $\sim 3 \times 10^8$. The pyramid shaped pedestal in the time range of $\sim 15\text{ps}$ close to the main peak has a relatively high intensity up to $\sim 3 \times 10^{-5}$ with respect to the main peak, corresponding to a coherent contrast of $\sim 3 \times 10^4$. There are also some sub-peak prepulses presented in the temporal profile, which will be discussed in the later section of this report.

For a designed intensity specification of Astra-Gemini laser at $\sim 10^{22}\text{W/cm}^2$, the incoherent ASE and coherent contrast of the existing laser system represent prepulse intensity at $\sim 10^{13}\text{W/cm}^2$ and 10^{17}W/cm^2 , respectively, which well exceeds the intensity threshold for plasma formation at $\sim 10^{10}\text{W/cm}^2$. Therefore, minimising the ASE background noise level was one of the major concerns of this investigation.

Various techniques have been investigated for possible improvement in the temporal quality. Firstly, a near-diffraction limited spatial filter has been implemented between the stretcher and the first amplifier (AMP1), in conjunction with a near-field aperture, to clean up any stray light into the amplifier. The measurement showed that there was no impact on the temporal contrast by either spatial filter or near-field aperture or both. Secondly, the extinction ratio of the fast pockel cell between the front-end and the stretcher has been optimised and improved by nearly one order of magnitude by using better quality polarisers. In addition, the original gold-coated back mirror in the stretcher was replaced by a dielectric-coated one to minimise losses and scattering due to the degradation of the

gold coating. Unfortunately, all these changes produced no observable impact on the contrast.

For an extreme power CPA laser system such as Astra-Gemini, the front-end provides the seed pulse for the subsequent amplifiers for further pulse energy amplification. Meanwhile the front-end has an amplification gain of $>10^6$, representing the highest gain section of the whole system. This implies that the ASE noise has been amplified the most in this section as well. Reducing the ASE noise level in the front-end system should therefore reduce the overall ASE background level of the whole system. To fulfil this task, the front-end system was upgraded by replacing the original JADE1 pump laser by a JADE2 for more reliable and stable operation. During this upgrade and system optimization, the Dazzler was relocated to a position after the first four passes of the multi-pass amplifier, and the Berek polarization compensator was removed altogether. The effect of these changes was to enhance the extinction ratio by a factor of up to 10, thereby significantly suppressing the intracavity ASE and enhancing the temporal contrast.

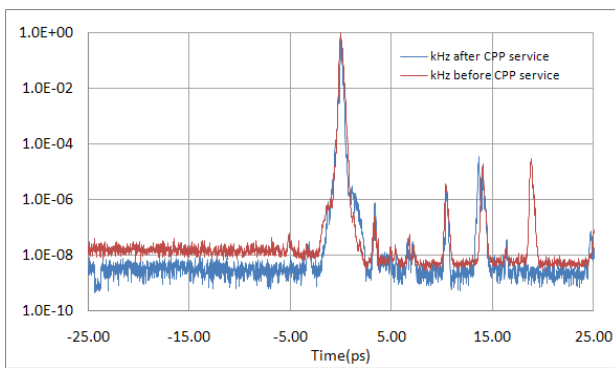


Fig.2 Front-end Contrast before and after upgrading

The output pulse from the front-end was directly recompressed to near transform limit by a prism compressor and the temporal contrast was measured with the Sequoia. Figure 2 shows the contrast measurement of the front-end before and after upgrading. It is clear that the ASE contrast has been improved by almost one order of magnitude. The contrast of the Astra-Gemini pulse was measured again after the front-end upgrading, and the result is shown in Figure 3. It is evident that the overall incoherent and coherent contrast of the Astra-Gemini laser pulse was enhanced by nearly one order of magnitude due to cleaner seed pulses delivered from the upgraded front-end.

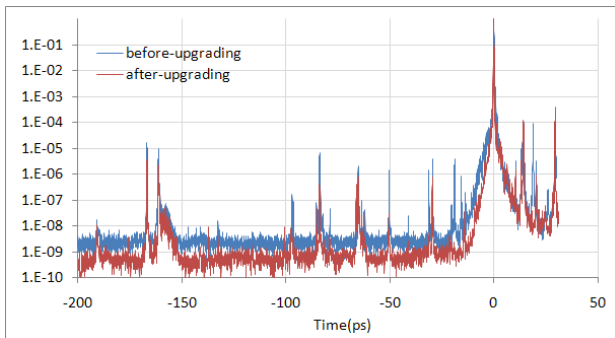


Fig.3 Overall contrast before and after front-end upgrading

As mentioned before and seen in Fig.1, there are a number of sub-peak prepulses appearing in the time range of up to ~ 200 ps prior to the main pulse. Unlike the incoherent and coherent pedestal prepulse, the sub-peak prepulses are replicas of the main pulse at much reduced intensity. They are much shorter than the ASE and coherent pedestals discussed above. The replica prepulses are usually generated by internal multiple reflections in various optical components, such as windows, lenses, polarisers and Ti:sapphire crystals, throughout the laser system. It is well-known that the post-pulse at a certain

temporal displacement with respect to the main pulse is generated at much reduced intensity by the internal Fresnel reflections even when the surfaces have antireflection coatings. The stretched, chirped post-pulse with a time separation from the main pulse less than the stretched pulse duration will be coupled with the chirped main pulse, producing both temporal and spectral modulation by interference. Any non-linear distortion of this modulation by high B-integral will generate new satellites after recompression. As a result, a counterpart prepulse of the post-pulse will appear before the main pulse when the pulse is compressed. The intensity of this prepulse is given by $\sim 0.6B^2W$, where B is the B-integral of the main pulse and W the intensity of the post-pulse [9]. Similar prepulses appear in contrast traces of many CPA systems, and are frequently (and wrongly) dismissed as artefacts of the measurement.

The replica prepulse can also affect the high field laser-matter interaction experiment dramatically by preheating or pre-ionizing the solid targets, and ideally should be suppressed and ultimately eliminated. To provide a cleaner and higher temporal quality pulse, a number of components in Astra with broadband antireflection coatings were replaced by optics having V-coatings, which typically have reflectivities of 0.1 to 0.2%, compared with around 1% for broadband coatings. By making these substitutions, some of the replica post-pulses and their counterpart pre-pulses were reduced by a factor of ~ 100 , as shown in Figure 4.

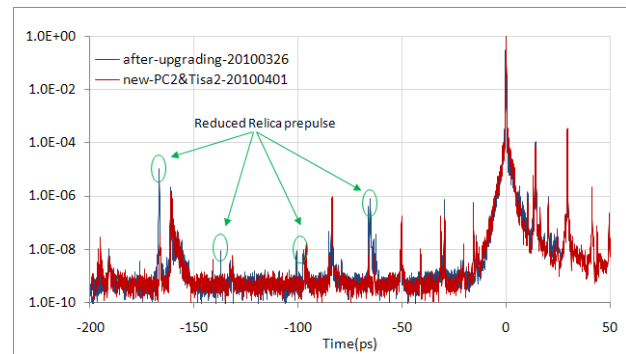


Fig.4 Replica pre-pulse reduction in the temporal profile

Conclusions

The temporal profile of Astra-Gemini laser pulse was extensively investigated under various conditions. The background contrast of the laser pulse was enhanced by nearly an order of magnitude by upgrading and optimizing the commercial front-end to provide a cleaner seed pulse. Some of the replica pre-pulses were reduced or eliminated by the use of components with better quality antireflection coatings.

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Electron transport during shock ignition

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Shock ignition (Betti et al, 2007, 2008; Ribeyre et al 2009) is one of the most promising routes to commercially viable inertial fusion energy (IFE). Compression is achieved on a low adiabat with minimum energy input. Once compressed, a strong shock is launched into the target by a high power laser. The shock converges on the centre of the target, heating the target and initiating burn.

We show here, with an idealized self-similar calculation, that laser interaction with dense targets during shock ignition can be separated into two regimes. In the ablative regime, corresponding to conditions found in conventional directly driven IFE, the interaction relaxes to a quasi-steady balance between electron thermal conduction carrying energy to high density and hydrodynamic energy flux (ablative enthalpy flow) which carries energy away from high density. In contrast, at high laser intensity, the heat flow Q into the target is too great to be balanced by hydrodynamic flow. In this regime, the structure is that of a supersonic heat front with very little hydrodynamic response.

In a calculation assuming that the Spitzer conductivity is acceptably valid, we show that conditions during shock ignition straddle the boundary between these two regimes. When reduced to its bare essentials, the laser-plasma interaction can be represented by 1D perfect gas fluid equations for mass, momentum and energy with additional energy fluxes carried by the laser and by Spitzer conduction (I and $Q = -\kappa \text{grad}(T)$ respectively).

To derive a self-similar solution to the fluid equations the dependence of the conductivity on the fluid variables is required. The Spitzer conductivity is determined by the mean free path λ of electrons as can be shown by writing the Spitzer heat flow as $Q = q(\lambda/L)Q_i$, where L is the temperature scalelength $L = T/\text{grad}(T)$. $Q_i = n_e k_B T (k_B T/m_e)^{1/2}$ is the free-streaming heat flow, and q is a numerical factor of order unity. For Coulomb collisions, λ is proportional to T^2 and this results in a self-similar solution which depends on the similarity variable $s = x/t^{4/3}$. ρ , u , P , Q & I can then be written as functions of s times a power of the ratio of time t to some characteristic time t_0 :

$$\rho = \rho(s) \quad ; \quad u = \mu(s) \left(\frac{t}{t_0} \right)^{1/3} \quad ; \quad P = \rho(s) \varphi(s) \left(\frac{t}{t_0} \right)^{2/3}$$

$$Q = - \left(\frac{\kappa_0}{t_0^{4/3}} \right) \varphi^{5/2} \frac{\partial \varphi}{\partial s} \left(\frac{t}{t_0} \right) \quad ; \quad I = \varepsilon(s) \left(\frac{t}{t_0} \right)$$

where κ_0 is a constant determining the magnitude of the Spitzer heat flow. ε is the self-similar laser energy flux. For laser energy absorption at a particular position s_{abs} , with a delta function energy input to the plasma where the density is ρ_{abs} , ε is a constant for $s < s_{\text{abs}}$, and $\varepsilon = 0$ for $s > s_{\text{abs}}$. The self-similar hydro equations in the overdense plasma ($I=0$) are then:

$$\frac{\partial}{\partial s}(\rho \mu) = \frac{4}{3} t_0^{1/3} s \frac{\partial \rho}{\partial s}$$

$$\frac{\partial}{\partial s}(\rho \mu^2 + \rho \varphi) = \frac{4}{3} t_0^{1/3} s \frac{\partial(\rho \mu)}{\partial s}$$

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$$\frac{\partial}{\partial s} \left(\frac{5}{2} \rho \varphi \mu + \frac{1}{2} \rho \mu^3 - \left(\frac{\kappa_0}{t_0^{4/3}} \right) \varphi^{5/2} \frac{\partial \varphi}{\partial s} \right) = \frac{4}{3} t_0^{1/3} s \frac{\partial}{\partial s} \left(\frac{3}{2} \rho \varphi + \frac{1}{2} \rho \mu^2 \right)$$

As expected, all quantities are now a function of one variable, s , instead of two, x & t . We find that not only are all quantities functions of s alone, but there is only one numerical constant, $\kappa_0/t_0^{4/3}$. This constant determines the ratio of energy flux by thermal conduction to energy flux by hydrodynamic motion and whether the solution lies in the ablative or heat flow regime. In the self-similar solution, the laser intensity rises linearly in time, and the rate of rise determines the numerical constant $\kappa_0/t_0^{4/3}$.

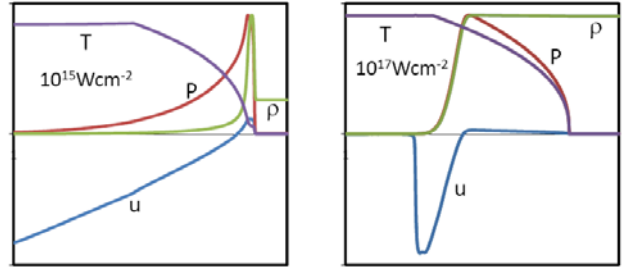


Figure 1. Profiles at 10^{15}Wcm^{-2} after 100ps and 10^{17}Wcm^{-2} after 1ps (arbitrary units)

Figure 1 shows how the laser-plasma interaction changes from the ablation regime at an absorbed laser intensity of 10^{15}Wcm^{-2} to a supersonic electron heat front at 10^{17}Wcm^{-2} . At both intensities the laser wavelength is $1/3 \mu\text{m}$ and absorption takes place at the critical density. The dense part of the target, at the right hand boundary, is initialised at 1 gm cm^{-3} . The laser intensity reaches the specified intensity at the end of a laser pulse of duration τ . The total laser energy is made the same at each laser intensity by choosing a pulse duration inversely proportional to the laser intensity such that in the left hand graph in the figure the intensity reaches 10^{15}Wcm^{-2} after 100psec, whereas in the right hand graph the intensity reaches 10^{17}Wcm^{-2} after only 1 psec. At 10^{15}Wcm^{-2} , the base of the temperature front coincides with the surface of the high density plasma. The high pressure launches a shock into the dense plasma and causes plasma to ablate from the surface. In contrast at 10^{17}Wcm^{-2} , the temperature front penetrates supersonically into the dense plasma with very little time for the plasma to start expanding from the surface.

Figure 2 plots the variation of the maximum pressure with laser intensity. At low intensity, the maximum pressure is proportional to laser intensity to the power 2/3 as expected for the ablation regime. At high intensity, the pressure is proportional to intensity to the power 2/9 as can easily be derived for a supersonic electron heat front. Figure 2 indicates that shock ignition lies at about the transition point between the two regimes, although the complicated density structures at the end of the compression phase must be taken account of for a realistic calculation, and non-local transport may affect the results.

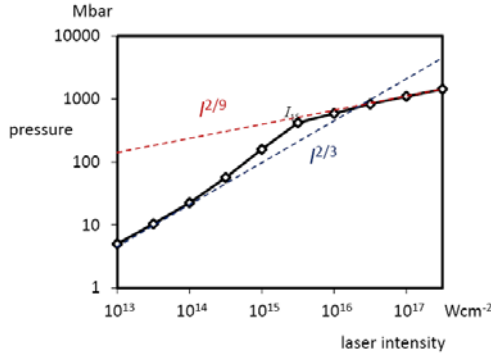


Figure 2. Pressure against intensity with the Spitzer conductivity

We assess the effect of non-local transport by time-dependent solution of the Vlasov-Fokker-Planck (VFP) equation self-consistently coupled to the perfect gas fluid equations. This amounts to replacing the Spitzer conduction equation with the VFP equation in our set of equations. The VFP equation for the electron distribution function $f(z, \mathbf{v}, t)$ in one planar dimension, z , is

$$\frac{\partial f}{\partial t} + \mathbf{v}_z \frac{\partial f}{\partial z} - eE \frac{\partial f}{\partial v_z} = C(f) + S(f)$$

where $C(f)$ is the collision term and $S(f)$ models laser absorption. The electric field is determined implicitly to produce zero electron current in the ion rest frame and maintain neutrality. The code uses the KALOS formalism (Bell et al 2006), which expands the distribution function in spherical harmonics. We find it sufficient for this problem to restrict the harmonic expansion to zeroth and first order, otherwise known as the diffusive approximation, which is equivalent to writing f as

$$f(z, \mathbf{v}, t) = f_0(z, |\mathbf{v}|, t) + f_1(z, |\mathbf{v}|, t) \frac{v_z}{|\mathbf{v}|}$$

Momentum is transferred from electrons to ions through the electric field and by collisions. The ion thermal capacity is smaller than electron thermal capacity by the ionization Z , here taken to be 4. Consequently we treat the ions as cold and ignore collisional energy transfer between electrons and ions. The electron distribution f is defined in the ion rest frame to simplify the collision term $C(f)$. The local ion velocity u varies in space. To allow for this, extra terms are added to the VFP equation as set out in equation 1 of Bell 1985. These terms are relatively small because the ion velocity is much smaller than the electron thermal velocity, but they are important because they include adiabatic cooling during rarefaction.

The dominant collision term is the angular scattering of electrons by ions which causes f_1 to decay exponentially. The electron-electron collision term takes the form of an advection-diffusion equation in momentum. The well-established approximation is made that the collision integrals, which are the equivalent of the Rosenbluth potentials, are calculated purely from the isotropic part of the distribution f_0 and a correction made to conserve momentum. The collision term is non-linear in the distribution function so we solve iteratively to conserve both electron number and electron energy to any prescribed accuracy down to rounding error.

At laser intensities of interest, collisionless absorption may be important, so energy is absorbed in a region surrounding the critical density by removing a fraction of the electron distribution each timestep and replacing it with an isotropic Maxwellian at a temperature $T_e + T_{\text{hot}}$ where T_e is the electron temperature (defined in terms of the mean electron energy) at the start of the timestep and T_{hot} is a hot electron temperature

specified in line with experimental data (Gitomer et al, 1986, Beg et al, 1997). The fraction replaced is chosen to give the required energy absorption.

We solve the VFP-hydro equations with a laser intensity rising linearly with time. This is equivalent to the self-similar calculation used above with the Spitzer-hydro equations except that it is not possible to begin the calculation with zero temperature or a step discontinuity in density at the critical surface. Nevertheless, the solution approaches the self-similar solution at the end of the calculation giving a meaningful comparison with the Spitzer-hydro results for the importance of non-local effects. For self-similarity the hot electron temperature T_{hot} varies in proportion to $t^{2/3}$, which makes $T_{\text{hot}} = \alpha I^{2/3}$ where α is chosen so that T_{hot} takes the required value at the end of the calculation.

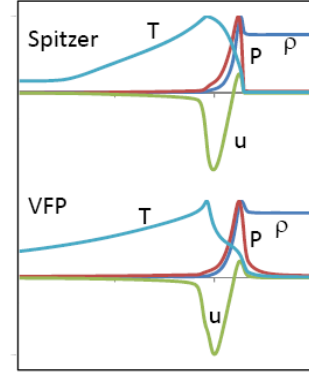


Figure 3. Spitzer and VFP ablation profiles at 10^{16} Wcm^{-2}

The profiles of temperature, velocity, density and pressure are shown in figure 3 for VFP and Spitzer calculations at an intensity of 10^{16} Wcm^{-2} . The VFP temperature profile is concave rather than convex. The pressure profiles differ little except that a VFP pressure precursor extends into the high density plasma due to the long mean free path of the more energetic part of the electron distribution.

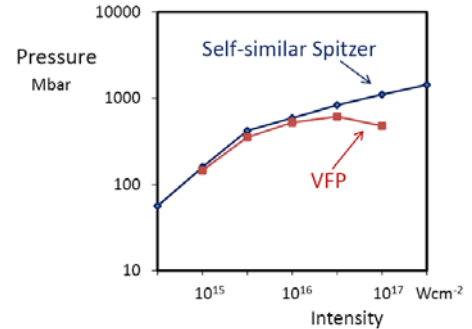


Figure 4. Pressure against intensity for VFP and Spitzer

As seen in figure 4, the VFP pressure only departs from the Spitzer pressure at high intensities well into the heat front regime. This is consistent with the conclusion from figure 3 that even if the VFP and Spitzer temperature profiles are different, the ablation process compensates for moderate intensities to give a similar pressure profile.

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New perspectives on the electrodynamics of intense laser-plasmas

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High-power lasers provide novel avenues for controllable investigation of matter in extreme conditions that only occur naturally away from the Earth. Moreover, it is expected that the next generation of ultra-intense lasers will, for the first time, allow controllable access to regimes where a host of different quantum electrodynamical phenomena will be evident [1]. The behaviour of intense laser-plasmas in such regimes is complex, and effective models will be indispensable for elucidating the influence of the quantum vacuum. In particular it is expected that semi-classical field theories, for example theories that include the Euler-Heisenberg Lagrangian [2], will play an important role and a key feature of such theories is that they are fundamentally non-linear. Virtual electron-positron pairs associated with the quantum vacuum provide an effective non-linear medium in which the electromagnetic field propagates.

Born-Infeld electrodynamics [3] has enjoyed a renaissance in recent years [4-7]. Unlike their Maxwell counterparts, the Born-Infeld field equations are fundamentally non-linear at the classical level; they are non-linear even in the classical vacuum. One can encode the vacuum Born-Infeld field equations in terms of non-linear constitutive relations expressing the electric displacement $\mathbf{D}$ and magnetic field $\mathbf{H}$ in terms of the electric field $\mathbf{E}$ and magnetic induction $\mathbf{B}$, leading to profound differences in the predictions of the Maxwell and Born-Infeld theories. In particular, Born and Infeld introduced their theory in order to ameliorate the singular self-energy of a classical point charge. However, it was also discovered that among the family of non-linear generalizations of Maxwell electrodynamics, Born-Infeld theory shares some highly attractive features with Maxwell theory; in particular, like the vacuum Maxwell equations, the vacuum Born-Infeld equations exhibit zero birefringence and its solutions have exceptional causal behaviour [8,9]. Moreover, Born-Infeld theory shares a number of properties with the low energy dynamics of strings and branes [10], and one can speculate that it may be more fundamental than Maxwell electrodynamics. If correct, this would revolutionize electrodynamics and have significant implications for the notorious problem of determining the classical force on a single accelerating point charge due to its own electromagnetic field, which has remained unresolved for over a century. Dirac [11] employed classical Maxwell electrodynamics, energy-momentum conservation and mass regularization to develop the Lorentz-Dirac equation of motion for an accelerating point electron which, unless special conditions are adopted for the final state of the electron, predicts that a classical free electron in vacuo can self-accelerate and the electron may experience a sudden acceleration before it enters a region of space containing a non-vanishing external electrostatic field.

As noted earlier, a host of different QED phenomena will be accessible to the next generation of intense lasers and non-linear electrodynamics in the context semi-classical QED will be central to their study. However, we speculate that Born-Infeld electrodynamics may play a role before it is necessary to invoke QED and this might become apparent in laser-plasma experiments over the next decade. Born-Infeld theory is characterized by a new fundamental constant of nature which limits the magnitude of the electric field of a static point charge.

To explore this further, we recently initiated a programme of work to elucidate the ramifications of Born-Infeld electrodynamics for high-field laser interactions with matter. In particular, we showed in [6] that a plane electromagnetic wave in a constant magnetic field propagates with a phase speed less than the speed of light in the vacuum and that this might be detectable in a terrestrial experiment. We also explored the behaviour of large amplitude electrostatic waves in a cold Born-Infeld plasma [7]. It is well-known that the amplitude of a plane longitudinal quasi-static oscillation of a relativistic cold Maxwell plasma is bounded from above (the so-called "wave-breaking" limit) [12,13], and the maximum electric field diverges as the phase velocity of the wave tends to the speed of light. We determined the corresponding maximum electric field in a cold Born-Infeld plasma and showed that it is bounded from above by the reciprocal of the Born-Infeld fundamental constant [7]. Furthermore, we extracted the Born-Infeld correction to the period of the wave in the ultra-relativistic regime of a cold Maxwell plasma.

In the high energy regime in which relativistic effects dominate – such as laboratory based laser-plasma acceleration – plasmas are commonly described by the collisionless Vlasov equation. An analysis of wave-breaking in warm Maxwell plasmas that directly employs distributional solutions to the Vlasov equation was given recently [14]. Alternatively, one may describe warm plasmas using the velocity moments of a one-particle distribution function. It is common to assume that the infinite hierarchy of field equations, obtained from velocity moments of the Vlasov equation, may be truncated to yield a closed set of field equations and a novel geometric averaging procedure for constructing velocity moments was recently developed [15-17] and used to establish rigorous conditions for the validity of the truncation in the ultra-relativistic regime. Recent work has also focussed on geometrical constructions of distributional solutions to the Maxwell-Vlasov system [18].

The collisionless approximation is often justified as the timescales governing relativistic processes in an underdense plasma are typically much shorter than the average time between collisions. However, recent advances in high energy density science have increased the demand for efficient descriptions of plasma dynamics fully incorporating both relativistic and collisional effects. Although the relativistic Fokker-Planck equation describes collisional plasmas in the relativistic regime, it contains a non-linear integral operator and is cumbersome to work with in many cases of interest. Furthermore, it does not easily lend itself to the generation of succinct fluid models. To address this issue, we recently developed [19] a relativistically covariant extension of the non-relativistic Lenard-Bernstein equation and used it to generate a new relativistic plasma fluid model that includes dissipative effects. Our induced fluid model contains new terms that arise from the non-trivial geometry of the unit hyperboloid in 4-dimensional (Lorentzian) phase space, and was used to investigate electric waves.

For small amplitude classical electromagnetic fields recourse is often made to a linear approximation scheme in which the appropriate constitutive relations arising from the coupled matter-field equations are linearized. If the medium is uniform

in space Fourier techniques are adequate since (if gravity is ignorable) the coupled system can be projected into plane-wave eigen-solutions of the Helmholtz equation. Such an approach leads to the concept of classical dispersion in which the parameters describing such eigen-solutions are required to satisfy constraints involving properties of the medium. However if the medium is inhomogeneous, exhibits relaxation or memory properties, or gravitational fields are present such Fourier methods no longer diagonalize the system and prove impractical. We have developed new approaches to circumvent these difficulties [20] and obtained integral equations that supercede classical dispersion relations in homogeneous media permitting investigation of Landau damping in non-stationary and inhomogeneous relativistic plasmas.

Finally, we have initiated investigation of the interaction of electromagnetic fields with accelerating matter by exploring constitutive relations for uniformly rotating media [21-23] and exploring how forces and torques can be defined covariantly and calculated for arbitrarily moving media. In particular, for a class of media with simple electromagnetic constitutive properties we show that, under the influence of an incident monochromatic, circularly polarized, plane electromagnetic wave, the Abraham and symmetrized Minkowski tensors induce different time-averaged torques on a uniformly rotating materially inhomogeneous dielectric cylinder and suggest that this observation may offer new avenues to explore experimentally the covariant electrodynamics of more general accelerating media.

In summary, work over the past twelve months in the Lancaster Mathematical Physics Group and Cockcroft Institute has led to a number of new avenues for exploring the behaviour of intense laser-plasmas.

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Cavitation and Shock Wave Formation in Dense Plasmas by Relativistic Electron Beams

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Introduction

Fast-ignition is an inertial confinement fusion scheme [1,2] where a separate igniter laser pulse is fired into a compressed capsule of deuterium-tritium fuel. This creates a beam of hot electrons which heat the compressed fuel close to the centre.

Once a high enough temperature is reached a self-sustaining burn-wave can propagate through the rest of the cold fuel and heat it up to fusion temperatures. The original fast-ignition scheme envisaged achieving this by using the igniter laser to bore up to the critical density in a plasma, where a beam of hot electrons would be produced that would heat the central area in the fuel [3]. Recently there has been more interest in cone-guided fast-ignition, where a hollow gold cone is embedded in the initial capsule, creating a clear path for the laser energy to reach the centre of the fuel [4, 5]. In either variant the propagation of a relativistic electron beam is a critical element.

A relativistic electron beam propagating through a background plasma will set-up a charge balancing resistive return current, such that

$$\mathbf{j}_f + \mathbf{j}_r \approx 0$$

where $\mathbf{j}_f$ and $\mathbf{j}_r$ are the forward going fast electron current and the return current respectively. A resistive electric field is formed such that $\mathbf{E} = -\eta \mathbf{j}_f = \eta \mathbf{j}_r$, where η is the resistivity of the plasma [6, 7]. This resistive return current will create a $\mathbf{j}_f \times \mathbf{B}$ force on the plasma and a pressure gradient via Ohmic heating, both of which would be expected to push the plasma away from the centre of the electron beam. 2D-3V Vlasov-Fokker-Planck simulations of this effect have previously been done by Kingham *et al.* [8], which showed collimation of the electron beam. A similar problem applied to cosmic rays passing through low density plasma has been looked at by Bell [9].

In this work we used a simple analytic model which predicts the density, pressure, magnetic field and velocity in the plasma to study the basics of cavitation in these circumstances. This model will be used to explore the strength of the cavitation over a wide range of parameter space that is of interest to fast-ignition fusion. Using an MHD code created specifically to look at this problem the analytic model will be shown to be effective at predicting the amount of cavitation that will occur in a

plasma. From both the analytic model and the full numerical simulation it is clear that the effect of the pressure gradient caused by Ohmic heating is dominant in the cavitation process. The region of hot electron current density and mass density can also be found where the pressure gradient from the Ohmic heating is so extreme that a shock wave can be launched into the surrounding plasma.

Theory

To understand what happens to the background plasma the collisionless fast electron current and resistive return current need to be added to the MHD equations. For Ampère's law we make a hybrid approximation of the form

$$\nabla \times \mathbf{B} = \mu_0(\mathbf{j}_f + \mathbf{j}_r) \approx 0$$

with the displacement current neglected. We also use an Ohm's Law of the form

$$\mathbf{E} + \mathbf{v} \times \mathbf{B} = -\eta \mathbf{j}_f$$

where there is a contribution to the electric field due to the resistive return current.

In addition the return current will cause Ohmic heating in the plasma

$$\frac{\partial T}{\partial t} = \frac{\gamma - 1}{n_i k_B} \eta j_r^2$$

where n_i is the number density of ions in the plasma, such that $n_i = \rho/m_i$, k_B is Boltzmann's constant and $\gamma = C_p/C_v$ is the adiabatic index. Throughout this work thermal conductivity is neglected.

Spitzer resistivity is used, which is given by

$$\eta(T) = 10^{-4} \frac{Z \ln \Lambda}{T_{eV}^{3/2}}$$

where Z is the ionic charge, $\ln \Lambda$ the Coulomb logarithm and T_{eV} the temperature of the background plasma in eV. This is valid for temperatures in the plasma above 100 eV at solid density, and is valid at lower temperature for lower mass densities. A fixed ionisation state is assumed. The return current is modelled as a rigid beam in 1D, with a Gaussian profile such that

$$j_r(x) = -j_f(x) = j_0 e^{-x^2/2R^2}$$

where R determines the radius of the electron beam.

These equations for the effects caused by the return current can be added into the MHD equations which describe the evolution of the plasma.

Although these equations are not able to be solved exactly analytically, by neglecting the advection terms in the resulting equations for the density, pressure, velocity and magnetic field we can obtain approximations for these parameters in the plasma, as a function of time and space. This allows us to take a parameter scan across the different values of hot electron current density and initial mass density to find the regimes where plasma cavitation is likely to be significant.

An example of the density profile in a hole boring fast-ignition scenario can be found in figure 3a of [10]. This shows that the fast electrons that are produced at the critical surface will have to cross a range of densities from 10 kg m^{-3} up to 10^6 kg m^{-3} . For much of this distance, hundreds of microns, the density is in the range $50 - 1000 \text{ kg m}^{-3}$. When comparing this range to figure 1 it can be seen that the cavitation effect will be significant over much of the distance the fast electrons are propagating over.

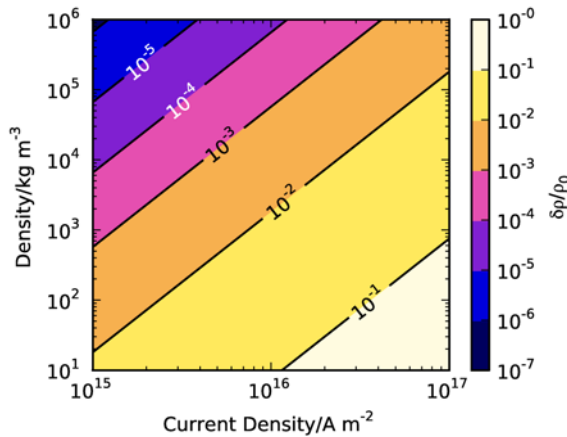


Figure 1. A parameter scan across density and current from the model. The initial parameters are $T_0 = 100 \text{ eV}$, $R = 3 \text{ } \mu\text{m}$, $Z = 1$ and $\ln \Lambda = 10$, and the final time is $t = 3 \text{ ps}$. The contour lines show increases of a factor of 10.

A similar treatment can be applied to find the parameters for which shocks are expected to form in the plasma. We have been able to obtain a formula for the velocity in the plasma from the MHD equations so it can be determined when $v_{\text{max}} > c_s$, where c_s is the sound speed in the plasma.

Figure 2 shows a parameter scan across the same values as in figure 1. The colour scale shows the time at which a shock would form in the plasma, which is at an initial temperature of 100 eV . At the top end of the colour scale times are $\geq 20 \text{ ps}$, meaning that no shock would be formed within 20 ps . The region in which the shock would be formed in $10 - 20 \text{ ps}$ is very narrow, as the transition is being made into the region where a shock would never form.

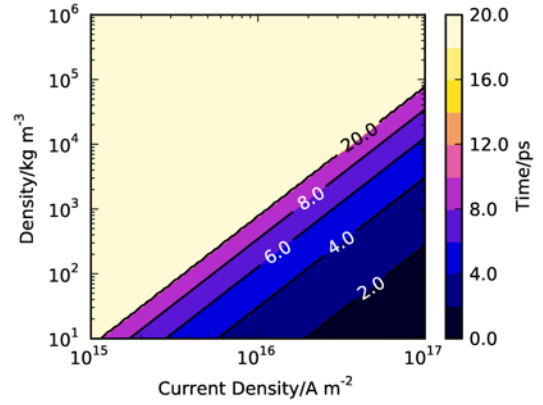


Figure 2. The time taken for shocks to form for different values of current density and mass density. The initial parameters are the same as in figure 1. Above the 20 ps contour the plot is saturated, so a shock would take more than 20 ps to form, if it would ever form.

Code Description

The 1D MHD code that was developed was based on the method by Ziegler described in [11]. This solves the MHD equations with the additional terms that arise due to the hot electron current.

The fast electron current is added into the code via the return current given in the previous section. Spitzer resistivity is used, along with the resistive magnetic field growth and Ohmic heating terms

As the problem here is only one dimensional some of the more sophisticated parts of the code, like the enforcement of $\nabla \cdot \mathbf{B} = 0$, are not required. Otherwise the implementation used here is as described by Ziegler, with RK2 integration of the MHD equations. Numerical diffusion is low due to the method of interpolation onto the cell walls, and open boundary conditions are used. The code was verified against the results of figure 3b in [12].

Results and Discussion

To begin with we will look at a typical case of interest to fast-ignition and compare the results from the code with the results predicted by the analytic model. In figure 3 the results are compared for the case where $j_0 = 10^{16} \text{ A m}^{-2}$ and $\rho_0 = 100 \text{ kg m}^{-3}$.

From figure 3 it can be seen that the analytic model and the MHD code give very good agreement. For the density the analytic solution over-estimates the cavitation in the central region, and underestimates it towards the edges. This can be understood by the lack of advection from the central region in the analytic model. The analytic prediction for the magnetic field is extremely close to the full numerical solution.

The plot for the velocity shows that the pressure gradient term alone gives a reasonable estimate of the velocity in the plasma. If a more complete consideration is used, including the $\mathbf{j}_r \times \mathbf{B}$ force, then the velocity becomes slightly underestimated in the regions where it is overestimated and vice-versa.

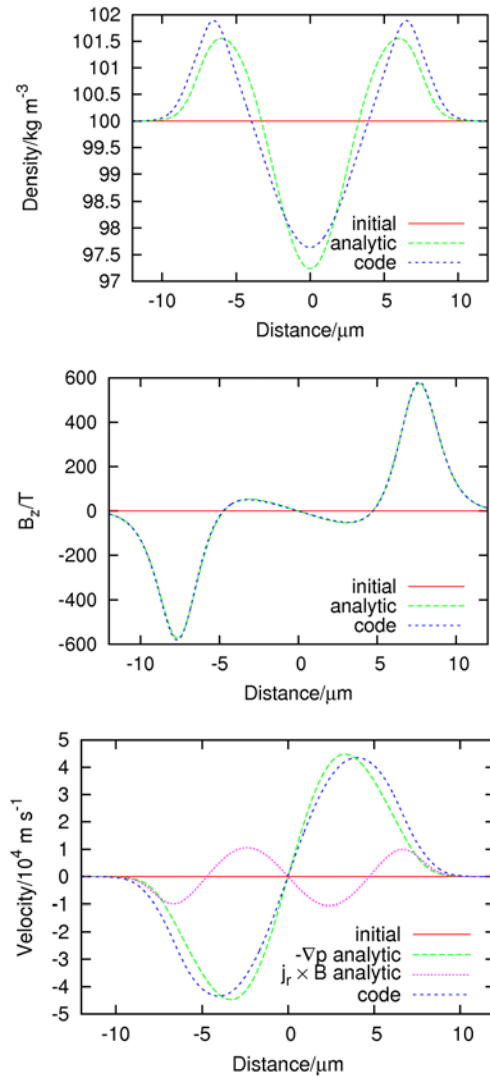


Figure 3. Plots of the density, magnetic field and velocity after 3 ps for $j_0 = 10^{16} \text{ A m}^{-2}$ and $\rho_0 = 100 \text{ kg m}^{-3}$. Otherwise the same values are used as in figure 1. In the plot for velocity the analytic contribution from the pressure gradient and $\mathbf{j} \times \mathbf{B}$ force are shown separately, demonstrating the dominance of the pressure gradient.

If the return current is driven for long enough at a high enough current density and low enough mass density shocks can form in the plasma. In figure 4 an example is shown for a return current of 10^{17} A m^{-2} and a mass density of 10 kg m^{-3} . At 5 ps the beginning of the shock formation can be seen and by 11 ps a sharp shock front has been formed. Because of the extreme amount of cavitation in this situation the analytic model can not be expected to give sensible results here.

From figure 4 it can be seen that we have essentially been able to produce a blast wave in this case. This is not unexpected, as we have rapidly put a lot of energy into a small volume. It should be noted that figure 2 shows only the time taken for $v_{\text{max}} > c_s$, and not the time for a well defined shock to form, as is only seen between 5 and 11 ps in figure 4. Not included in the MHD code is the effect of radiative heating, in this regime it will certainly have an effect on the form of the shock front obtained.

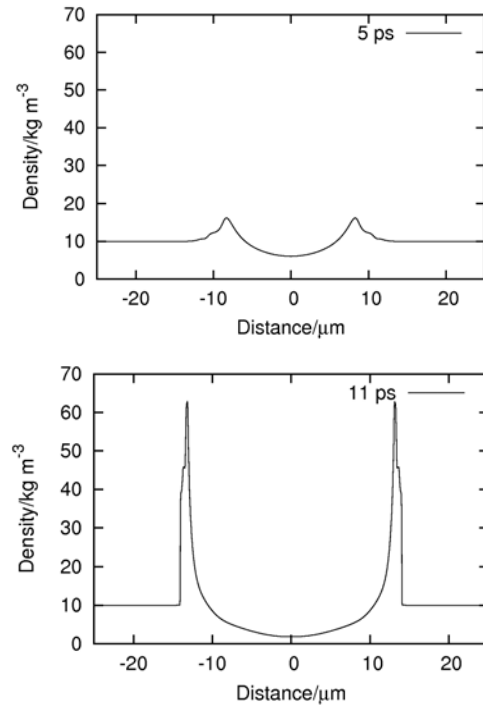


Figure 4. Density plots from the MHD code showing shock formation caused by the resistive return current. The initial parameters in this case were $j_0 = 10^{17} \text{ A m}^{-2}$ and $\rho_0 = 10 \text{ kg m}^{-3}$, other parameters were the same as for the results shown in figure 3.

Conclusions

It has been shown that for parameters of interest to fast-ignition the cavitation effect can be quite significant, getting up to 10% of the initial density after just 3 ps. The good agreement between analytic theory and fully non-linear MHD simulations indicates that one can be confident that cavitation is relevant to fast-ignition. Similarly shocks can be formed in fast-ignition relevant plasmas after just a few ps. These were predicted by the analytic model and an example from the MHD code has been shown.

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Alpha-particle stopping power for dense hydrogen plasmas

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Introduction

With the completion of the NIF facility, inertial confinement fusion (ICF) has made considerable advances and can now be considered as a promising method for an energy source for the future [1,2]. Achieving ICF involves the creation of a dense, high temperature plasma. The way in which charged particles pass through such plasmas and deposit their energy is of interest to ICF research for two primary reasons. The first case is in the simulation of ion beams as primary drivers in ICF [3]. In this case, knowledge of the distribution of the beam energy across the target is important in order to reduce the probability of instabilities arising.

The second case is the modeling of the α -particle heating experienced by dense plasmas surrounding the hot spot where fusion reactions are already taking place. Immediately after the initial, externally driven, compression stage during ICF experiments, sufficient temperature and density conditions for fusion [4] are created at the centre of the fuel capsule. The fusion reactions that occur create α -particles that deposit their kinetic energy in the surrounding plasma and heat a part of the outer highly compressed plasma to fusion temperatures. As a result more α -particles are created from new fusion reactions. This dependent chain of fusion reactions and further heating is a burn wave that needs to propagate from the centre of the fuel outwards through the whole capsule, to allow for efficient, high gain targets. Thus, the energy deposition in high-density hydrogen is important to describe the creation of a burn wave and, thus, modeling conditions for high gain targets.

To simulate the energy deposition process, we calculate the stopping power as a rate of kinetic energy lost by an incident particle per unit distance traveled through the stopping medium, $-dE/dx$. Numerically integrating this quantity along the beam path will yield the energy deposition profile of the α -particles. Different beam profiles can easily be generated by weighted distributions of initial energies.

In this work, we combine advanced kinetic models for the beam-plasma interactions [5,6] with state of the art descriptions of the target plasma. The latter consists of dense hydrogen and is described by full quantum simulations (DFT-MD) [7-10]. A one-dimensional density line profile is then calculated by taking the density along a line through the simulation box at an oblique angle. This profile is used as a dynamic input parameter for the calculation of the energy loss of α -particles with an energy of 3.5 MeV passing through this plasma.

Many existing models currently describe the stopping power experienced by ions in a variety of target mediums. Dense plasma effects such as quantum degeneracy, self-energy, ionisation and dynamical screening are not taken into account in more classical stopping models [11,12]. Accuracy of existing models can be improved upon by recalculating the stopping power using a quantum mechanical description of the system and the creation of High Energy Density (HED) states of matter allows experimentation in new regimes of matter where models incorporating these effects can be tested.

DFT-MD Simulations for the Target

The density profile of the hydrogen target is calculated through quantum density functional molecular dynamics (DFT-MD) simulations using the simulation package VASP [8-10]. From an initial density, a bcc crystal structure of hydrogen nuclei is produced. The number of ions necessary to form the periodic DFT system is determined by the particular crystal structure used and the spatial size of the system is determined by the initial density. These ions are then given an initial Maxwellian thermal velocity distribution. The electrons are initialized, often as atomic orbitals.

From this converged electronic configuration, in the external field of the ions, new forces on the ions can be calculated. With these, a molecular dynamics step of the ions is performed. The resulting new ion configuration gives rise to a new solution of the Kohn-Sham equations governing the electron configurations and, thus giving a new electronic density configuration. This can be used to implement the next MD step.

The exchange correlation functional, describing electron interactions, is the major ingredient of the DFT step for the electrons. It modifies the potential felt by the electrons due to the many-body interactions and Pauli blocking. As this potential depends on the electron configuration, convergence must be reached between the new electron wave functions and the exchange correlation functional. Throughout the simulation the ion temperature is kept constant using a Nose-Hoover thermostat and the iterative DFT-MD process is repeated until the lowest free energy of the electrons is reached (see Ref. 13).

Using this method, a periodic three-dimensional density profile is formed, which includes both the locations of the nuclei and the electron density at cubic grid points. Then we start from a random footpoint in the DFT box and follow a path whose direction is set by random angles of azimuth and inclination. In this way, a one-dimensional line density can be extracted. To calculate the density at a generalized co-ordinate a trilinear interpolation scheme is applied to the densities at the eight neighboring grid points of the box. The profile is evaluated at 0.1 Angstrom increments along the described path. Fig. 1 shows an example of a line profile obtained by this procedure.

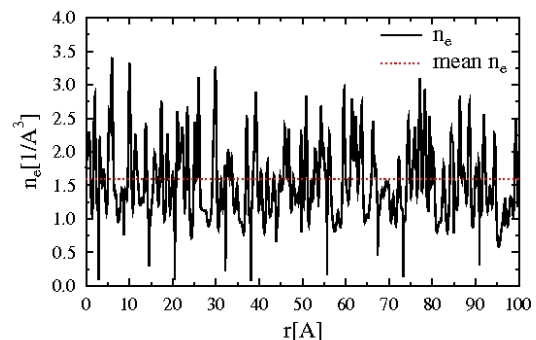


Fig. 1: Line density profile segment taken from a DFT-MD simulation for dense hydrogen with $T=10000\text{K}$ and $n_e = 1.661 \times 10^{24}\text{cm}^{-3}$.

Quantum Calculations of Stopping Power

The stopping power experienced by a charged particle traveling through a plasma, which is due to elastic binary collisions, can be calculated by the following expression which follows from the quantum Boltzmann equations [14,5].

$$\begin{aligned} \frac{\partial}{\partial t} \langle E \rangle_{T-MD} = & \frac{1}{(2\pi)^3} \sum_{\mathbf{p}} \frac{m_e^2}{m_e^2} \frac{n_e A_{\mathbf{p}}^2}{p} k_B T \int_0^\infty dp p^2 Q_{\mathbf{p}}^T(\mathbf{p}) \\ & \times \left[\left(1 - \frac{m_{De} k_B T}{m_e p} \right) \exp \left[-\frac{m_e}{2k_B T} \left(\frac{p}{m_{De}} - v \right)^2 \right] \right. \\ & \left. + \left(1 + \frac{m_{De} k_B T}{m_e p} \right) \exp \left[-\frac{m_e}{2k_B T} \left(\frac{p}{m_{De}} + v \right)^2 \right] \right]. \end{aligned}$$

The evaluation of this expression requires the momentum transfer cross section for electron-ion scattering Q^T . This cross section can be determined from phase shift analysis of the electron wave functions [15], which are dependent on the potential through which the incident beam is scattering. Here, we use a Debye potential to take screening into account. The complex scattering for a charged particle passing through a large composite body is here reduced to multiple two-body scattering events.

To calculate the phase shift the Numerov algorithm [16] is employed to solve the radial Schrödinger equation,

$$\left(\frac{\partial^2}{\partial r^2} + E - V(r) - \frac{l(l+1)}{r^2} \right) \psi(r) = 0$$

The phase shift of a scattering state is the measured difference in phase between the wave function as calculated in the potential, $V(r)$, and the wavefunction as calculated for free electrons. Phase shift analysis [15] of scattering states, over all available linear and angular momentum values, allows calculation of the momentum transport cross section via [13]

$$Q^T = \frac{4\pi}{k^2} \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_l(k) - \delta_{l+1}(k))$$

Stopping power in binary collision approximation can then be calculated by substituting the calculated momentum transport cross sections into the Boltzmann expression. Dynamic screening effects can be modeled by using a velocity-dependent screening length [5,6], which also accounts for the excitation of collective modes (plasmons). For this work, the fit proposed in Ref. 10 has been employed in order to reduce the computational intensity of calculating this stopping power.

Energy Loss Comparisons

A solver has been written which takes inputs of temperature and density, calculates the stopping power and simulates the energy loss of an α -particle as it passes along a one-dimensional path.

Bragg curves, stopping power versus stopping path length, are calculated by the solver for 3.5 MeV α -particles. These curves are compared for the cases of stopping calculated using the DFT-MD density profile and for stopping power calculated using the mean density of the three-dimensional dense hydrogen DFT-MD simulation box.

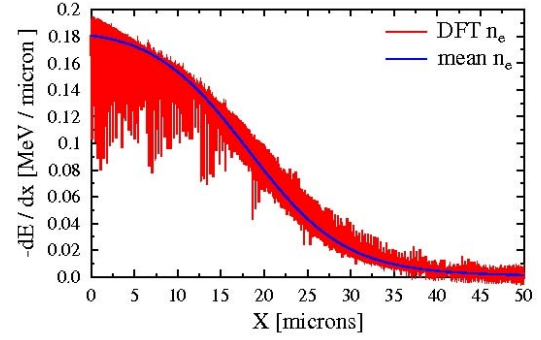


Fig. 2: Comparison of α -particle stopping in hydrogen using a density profile obtained by DFT-MD and a constant mean density. The beam energy is $E = 3.5\text{MeV}$. The plasma has a temperature of $T=10000\text{K}$ and a mean electron density of $n_e = 1.661 \times 10^{24} \text{cm}^{-3}$.

Despite the frequent large fluctuations in the density profile, causing large fluctuations in the calculated values of stopping power, there is good agreement between the general functional form of the Bragg curves for the two cases and also between the ion ranges calculated in the two cases. This implies that, for a beam path length much greater than the width of the DFT profile, the energy deposition profile of the incident particles can be well approximated by calculations that use the average target density. However, the straggling of the beam is strongly underestimated by considering a constant mean density as the quantum simulations show huge microscopic fluctuations that are fully reflected in the Bragg curves.

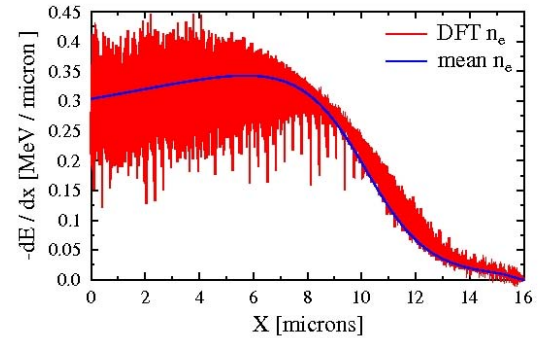


Fig. 3: As Fig. 2, but for a temperature of $T=20000\text{K}$.

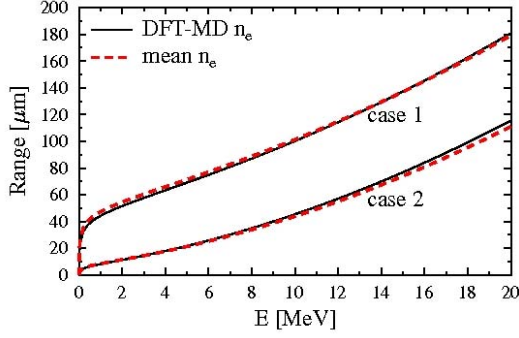
Range Comparisons

The ion range is the distance at which all of the kinetic energy of the ion has been deposited in the stopping medium. Actually, the ion is not fully stopped at this point, but thermalized to the plasma ions. The depth of this point into the stopping medium is obviously highly dependent on the initial kinetic energy of the incident particle and the plasma conditions.

For a well-defined analytical solution for the stopping power, the range can be calculated from the integral

$$Range = \int_{E_0}^0 \left(\frac{dE}{dx} \right)^{-1} dE$$

However, the ranges have been calculated in this work using the stopping solver to integrate along the beam path until the step at which $E_B \leq 0$, where E_B is the spatially dependent energy of the beam particles.



Range of α -particles in dense hydrogen using the highly fluctuating electron density from DFT-MD simulations (see Fig. 1) and a constant mean density. See Figs. 2 and 3 for the plasma conditions (in particular the temperatures) of cases 1 and 2, respectively.

The α -particle ranges are calculated, at initial kinetic energies from zero to 20 MeV for both mean and fluctuating electron density from DFT-MD. For the case of the beam velocity being less than the electron thermal velocity in the target medium, the stopping power can be approximated as a linear frictional force, $-dE/dx = Bv$. Solving for the analytical solution to the range in the low energy approximation shows good agreement with the numerical calculations. In the high velocity case, the Bethe limit can be employed and the stopping power is approximated by $-dE/dx = C/v^2$. It is also found that analytical solutions of the range solved from this form of stopping power show good agreement to the ion ranges calculated by the numerical solver. The comparison in Fig. 3 demonstrates that the range of the α -particles can be calculated from mean densities as the large fluctuations in the density cancel during the stopping paths. Thus, the predicted size of the hot spot needed in ICF targets is mainly determined by the physics included in the stopping power model employed. More advanced models [5,6,18,19] should receive more attention although they are numerically difficult to implement into a hydrodynamic simulation.

Conclusions

For beam energies from 0–20 MeV, a good agreement is obtained between calculated ion ranges for α -particles stopping in dense hydrogen for both highly fluctuating electron densities from DFT-MD and uniform averaged density profiles. In addition, the range calculations demonstrate good agreement with analytical range calculations using stopping power approximations in the limit of both the high and low kinetic energy as well.

This agreement implies that for calculations of large-scale effects, such as ion range or averaged energy deposition, the increased accuracy provided by applying a DFT-MD density profile has little effect on the final result. It is also observed that the general functional form of the Bragg curves calculated for the variable and uniform density cases is similar. However, in variable density DFT-MD cases, fluctuations in density cause dramatic changes in the calculated stopping power over small spatial scales. This huge straggling must be considered in situations with small fluxes where heating cannot be averaged over many particles.

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An Object-Oriented 3-D View-factor Code for Hohlraum Modelling

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Introduction

The design of the hohlraum plays a crucial role in indirect-drive ICF. Both the hohlraum geometry itself and the configuration of incident drive lasers are major factors in determining whether an experiment will be successful. Alteration of these factors will alter both the driver-capsule coupling efficiency and the uniformity of implosion of the fuel capsule. Analytic approaches can be utilized to give a theoretical insight into these effects, but often require limiting simplifications such as a spherical hohlraum^[1]. Alternatively, a numerical approach can be used to obtain a more realistic model of the system.

A radiation hydrodynamics simulation would deliver the best predictive results in this situation. While accounting for physical processes on a fine level of granularity, these calculations can be prohibitively computationally expensive; especially if performed in three dimensions. In addition, models can be difficult and time consuming to correctly set up. Less complex are view-factor methods. These allow a whole hohlraum to be modelled relatively inexpensively, although low level details (such as driver-wall coupling) must be parameterised rather than calculated *ab initio*^[2].

View-factor based hohlraum modelling codes use simple geometric arguments to determine the quantities of radiation that can flow to one part of a hohlraum geometry from another, and then solve a power balance equation to calculate the spatial distribution of radiation throughout a target^[3]. Because of the relative speed involved in setting up and running simulations, 3D view-factor codes are especially suited to prototyping experimental target designs.

While very useful for experiment design, existing commercial codes can be costly and may also represent a “black box” since source code may not be provided. Thus, we created a new, easy to use 3D view-factor code that can be employed to study radiation effects in a wide range of hohlraum geometries. It can be used to easily create hohlraum geometries from a number of geometric primitives. Drive laser configurations can be specified, and are implemented via a ray-tracing algorithm. Given a pulse shape, the code can then calculate the time-dependent evolution of the radiation distribution throughout the hohlraum. As well as raw data output, visualisation tools have been created to display the resulting data as a 3D false colour animation.

Mesh Generation

For numerical techniques to be applied, a hohlraum model must be discretised into a mesh of triangular faces. Triangles as a basis for mesh construction are preferred for a number of reasons. The first is that triangle vertices are guaranteed to be co-planar – a triangle's normal cannot vary over the face. This is important as face normals are used heavily in the calculation of the view-factors. Second, simple and efficient tests exist for an intersection between a line and a triangle, which aids computation speed when implementing ray-tracing and occlusion detection algorithms. To simplify calculation, faces have normals in one direction only; they are effectively transparent from the back.

The user can construct hohlraums by combining cylinders, disks, annular rings and spheres. The dimensions of these objects, their orientations and positions are specified in an XML configuration file. As an aid to the process, a designer application is included that allows the user to generate the configuration files in a more user friendly manner. As well as being capable of creating and editing the properties of various primitives, the resulting mesh can be viewed and rotated in a 3D preview window (See fig.1).

The *meshGenerator* program takes a configuration file created by the designer as a command line argument, and writes a binary file containing the face data. This file can be used as input to the other programs in the code, and contains the positions, areas and normals of each face.

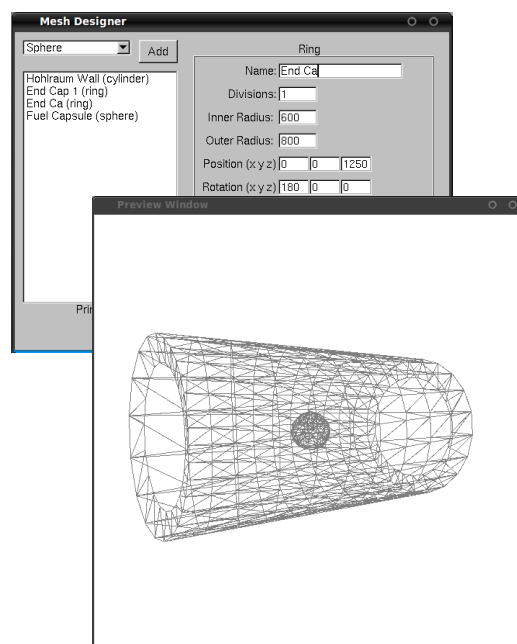


Figure 1 – The mesh designer application is a graphical interface that allows the user to easily create model configuration files for the code by interactively editing properties of 3D primitives.

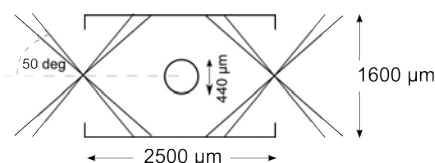


Figure 2 – The dimensions of the hohlraum used in the HEP-1/Precision Nova experiments at LLNL [8]. This series of experiments were numerically modelled to determine if the code could successfully reproduce experimental results

View-factor Calculation

Once all geometric details of the mesh have been calculated, the possible ways in which radiation can flow from one face to another may be evaluated. This is accomplished by means of a 2D view-factor matrix. The view-factor between two faces F_{ij} is the fraction of total energy that leaves face i and arrives at face j . This is given exactly by the view-factor integral^[4]:

$$F_{ij} = \frac{1}{2\pi A_i} \int_{A_i} \int_{A_j} \frac{\cos \theta_i \cos \theta_j}{\pi S^2} dA_i dA_j$$

Where A_i and A_j are the areas of faces i and j , S is the distance between the differential area elements, and θ_i and θ_j are the angles between the normals of the respective elements. In the limiting case of a large number of small, co-planar faces this equation can be simplified to

$$F_{ij} = \frac{1}{\pi A_i} \frac{A_i A_j}{S^4} (\hat{n}_i \cdot \vec{r})(-\hat{n}_j \cdot \vec{r})$$

Where $\hat{n}_i$ and $\hat{n}_j$ are the normal vectors of faces i and j respectively, and $\vec{r}$ is the vector from face i to face j . The reciprocity relation $A_i F_{ij} = A_j F_{ji}$ saves us from having to calculate the entire matrix; once F_{ij} is known F_{ji} can be trivially calculated.

The calculation is complicated by the possibility of a view-factor being totally or partially occluded by another face. In this implementation, we assume if the vector between the centre of two faces intersects any other face in the mesh, the view-factor between those faces becomes zero. This becomes a more accurate approximation as mesh resolution is increased, but at the same time introduces a significant computational overhead. If required for every face in the mesh, the complexity class of this algorithm becomes $\sim O(N^3)$ for N faces; as such the calculation of this matrix is generally the most time consuming part of the computation. The view-factor matrix is generated in this code by the *viewFactorCalc* program, which takes a mesh file as its input, and writes out a file containing the view-factor matrix.

The implementation of the view-factor calculation in this code was tested by comparing its results to analytically known solutions. A classic example is the view-factor between two circular parallel plates. Analytically, the view-factor between two parallel circular plates of radius $1000\mu\text{m}$ with a separation of $1500\mu\text{m}$ is 0.25^[5]. With a plate mesh surface resolution of 802.1 mm^2 , the numerical error was 4.6 parts per million – a small error within manageable low resolutions.

Beam Ray-tracing

A ray-tracing algorithm was used to allow a realistic representation of a set of lasers. A laser can be described by a focal point, a direction vector, and an F-number. Once these quantities are known, the beam can be approximated by a large set of evenly spaced rays, each carrying an evenly distributed fraction of the beam's total power. The power of the ray is then applied to the intersecting face closest to the laser source. In this initial version of the code, the coupling between beam and surface is assumed here to be perfect – all the power from each ray is re-emitted diffusely by the face it is incident upon. This is a fair approximation, despite the complex physics involved in the x-ray generation process: depending on the target design, a large fraction of incident energy can be converted to x-rays^[6].

In order for this to produce realistic results, the number of rays in each beam must be such that many rays fall on each face. This is achieved by automatically increasing the density of rays in each beam until the average number of rays intersecting with a face is at least ten. This ensures a smooth and realistic distribution of beam power on arbitrary resolution geometries.

Drive beam configurations are also specified in an XML configuration file, along with a few other simulation parameters. As well as creating individual beams, the user can also easily create rings of beams by additionally describing the number of beams in each ring and their angle to a direction vector.

Solving the Radiosity Equation

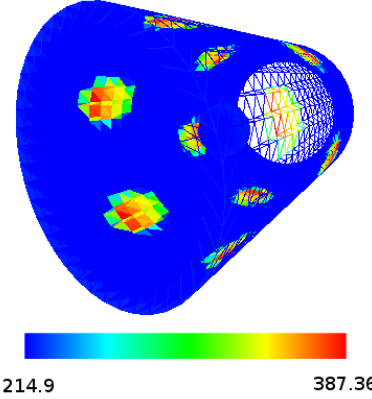


Figure 3 - Numerically calculated distribution of radiation temperature in eV over the surface of the HEP-1 target. This image was taken from the 3D viewer developed for use with the code.

In order to calculate the static power balance condition throughout a hohlraum mesh, one can consider the radiosity equation, which must be true in a steady state^[7]:

$$B_i - \alpha_i \sum_j F_{ij} B_j = E_i$$

Where B_i , B_j are the radiosities of faces i and j , α_i is the albedo of face i , and E_i is any external power contribution to face i – e.g. power delivered from a laser. The radiosity of a face is the power it emits per unit area. By Stefan-Boltzmann law, it may be expressed as $B_i = \sigma T_i^4$, where T_i is the face temperature and σ is the Stefan-Boltzmann constant. The radiosity equation is an expression of the fact that in a steady state, the power leaving a face must be equal to the power incident on the face. The only unknown parameter in this equation is the albedo; here it is simplified to a constant scalar value. Later it will be seen that this is a reasonable approximation.

Using matrix inversion methods exact solutions to this equation can be found, but this can be computationally expensive; additionally, for most cases exact solutions are not required. In this code, a Jacobi iteration is used to converge upon the solution. Once a user defined minimum percentage difference between old and new radiosity values has been met, the solution is deemed good enough and the process terminates. The final level of convergence can also be used to estimate the error in the numerical result.

The radiation transport code was tested by comparing its numerical results to experimentally observed hohlraum temperatures. The Nova laser at Lawrence Livermore National Laboratory conducted widely-documented experiments with various hohlraum geometries, dummy capsules and laser powers^[8]. Here, the HEP-1/Precision Nova series of experiments were selected for modelling - the dimensions of the hohlraum used can be seen in fig 2.

A suitable XML configuration file to describe the experiment's hohlraum was created and used as input to the *meshGenerator* program. The view-factors were then calculated. This particular experiment utilises two beam cones at an incident angle of 50 degrees to the axis – each has five beams, and the peak power

of the pulse is 18TW. The radiation transport code was run with these parameters to a convergence level of 0.1% - the results of the radiation distribution around the interior of the hohlraum can be seen in fig. 3.

Because the albedo is the only remaining degree of freedom in the problem, comparison to experimental data allows the albedo to be calibrated to an effective value. In this case, a simulation albedo of 0.8 yielded a calculated fuel capsule surface temperature of $230.1 \pm 2.7 \text{ eV}$. The measured peak temperature from the Nova experiment was determined to be $227 \pm 3 \text{ eV}$ from fig. 4 - this suggests that 0.8 appears to be an appropriate value for the effective albedo.

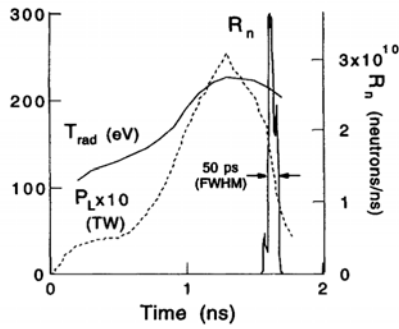


Figure 4 – Experimentally measured hohlraum temperatures in the HEP-1 experiments, reproduced from ref. 8. Also shown is the experimentally measured drive laser ps22 pulse shape.

Modelling the Temporal Evolution of Hohlraum Temperature

After it was known the code could produce realistic results for a given steady power input, it was altered in order to calculate time-dependent radiosity distributions. This is a reasonably straightforward extension of the previous methods. Time-dependence becomes important when the effects of laser pulse shaping are studied. As such, a method of specifying arbitrary pulse shapes and powers was needed. In this implementation, one or more pulse shapes can be specified independently in a transport configuration file. Each beam can then refer to a named profile as it is created.

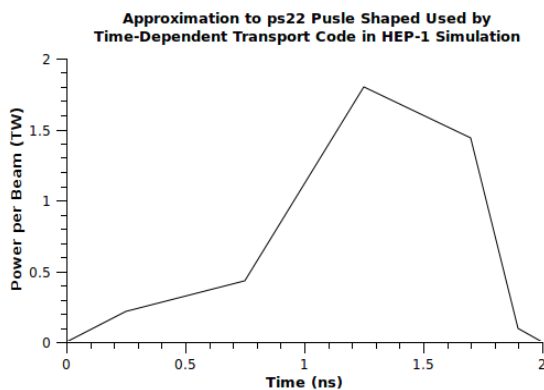


Figure 5 – The approximation to the ps22 pulse shape used in the time-dependent simulation. The shape is defined by a set of points; linear interpolation between the points is used to determine the appropriate beam power at a given time step.

The user defines the pulse shape by supplying a list of points. Each point defines the power of the beam at a given time. By building a list of points in this way, increasingly better approximations to the desired power profile can be created. The time-dependent program then linearly interpolates between the points to determine the applied beam power for each time step. The radiosity balance at each point in time is then solved in the

same manner as before, and the distribution of radiosities is saved for each time step.

As a comparison, the same Nova HEP-1 model was extended by defining a time dependent pulse shape. In the Nova shots, the named ps22 pulse shape was used^[9]. The approximation to this pulse shape used is shown in fig. 5, and the resulting simulated capsule surface temperatures are shown in fig. 6.

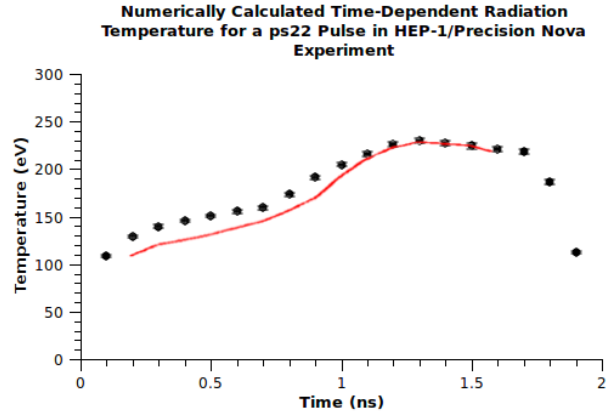


Figure 6 – Time dependent radiation temperature calculation compared with experiment. Points are numerically calculated temperatures over the fuel capsule, with error bars representing non-uniformities. The continuous line is the experimentally measured temperature rescaled from fig. 4.

It can be seen that this is quite a reasonable representation of the experimental results. The form is closer to experimental values around the peak temperatures as this is the region that the albedo was calibrated for; although even for lower temperatures (around 100eV) the divergence from experimental values is quite minimal. This might suggest that the albedo does not vary a great deal over this range of temperature, and that a single value albedo is a quite reasonable approximation. An improved approximation to the ps22 pulse shape might also yield improved results.

Conclusions

This project has developed a basic but capable and user-friendly 3D view-factor code for use in designing high-power laser experiments. It has the ability to model a wide range of user-defined geometries, calculate the appropriate view-factors, and then solve the radiation transport equation in a time-dependent or time-independent manner. It also provides 3D visualisation utilities that can be used to view the results of these operations.

The view factor calculation algorithm has been well tested. Analytically known view-factors can be solved with errors of only a few parts per million using manageably low resolutions. Occlusion is successfully handled, but comes at the price of increased algorithmic complexity. One way to speed up calculation of the view-factor matrix is to implement this as a parallel computation; and this should be relatively straightforward given the nature of the algorithm.

The radiation transport section of the code performed well, despite much of the complexity associated with the underlying physical processes being parameterised in the radiosity equation. It has been shown to reproduce experimental results quite well; as such, it should be able to provide significant insight into the design and performance of new hohlraums for high power laser experiments

Acknowledgements

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Siegert states in an electric field: From atoms to molecules

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1. Introduction

In a previous work [1], within the single active electron approximation, the Siegert states of atoms in a static uniform electric field, defined as the solutions to the stationary Schrödinger equation satisfying the regularity and outgoing-wave boundary conditions [2], are discussed. In [1], an efficient method to calculate not only the complex energy eigenvalue, but also the eigenfunction for a general class of one-electron atomic potentials is introduced. An exact expression for the transverse momentum distribution of the ionized electrons in terms of the Siegert eigenfunction in the asymptotic region is derived.

In this report, we present an extension of the Siegert states to molecules. The formulation of the computational method in this case requires some modifications.

The Hamiltonian for an electron interacting with a model molecular potential $V(\mathbf{r})$ and a static uniform electric field $\mathbf{F}$ directed along the z axis has the form (atomic units are used throughout)

$$H = -\frac{1}{2}\Delta + V(\mathbf{r}) + Fz. \quad (2)$$

We use the parabolic coordinates ξ , η , and φ defined as follows:

$$\xi = r + z, \quad 0 \leq \xi \leq \infty, \quad (3)$$

$$\eta = r - z, \quad 0 \leq \eta \leq \infty, \quad (4)$$

$$\varphi = \arctan \frac{y}{x}, \quad 0 \leq \varphi \leq 2\pi \quad (5)$$

In [1] we have considered the case when the potential has axial symmetry, $V(\mathbf{r}) = V(\xi, \eta)$. Then the solution of Eq. (1) can be sought in the form

$$\psi(\mathbf{r}) = \frac{e^{im\varphi}}{\sqrt{2\pi}} \psi(\xi, \eta), \quad (6)$$

2. Basic equations

The Schrödinger equation reads

$$H\psi = E\psi. \quad (1)$$

where m is the magnetic quantum number. The function $\psi(\xi, \eta)$ satisfies

$$\left[\frac{\partial}{\partial \eta} \eta \frac{\partial}{\partial \eta} - \frac{m^2}{4\eta} + \mathcal{B}(\eta) + \frac{E\eta}{2} + \frac{F\eta^2}{4} \right] \psi(\xi, \eta) = 0, \quad (7)$$

where the adiabatic Hamiltonian $\mathcal{B}(\eta)$ is given by

$$\mathcal{B}(\eta) = \frac{\partial}{\partial \xi} \xi \frac{\partial}{\partial \xi} - \frac{m^2}{4\xi} - \frac{\xi + \eta}{2} V(\xi, \eta) + \frac{E\xi}{2} - \frac{F\xi^2}{4}. \quad (8)$$

These equations must be supplemented by the regularity and outgoing-wave boundary conditions [1]. For non-zero electric fields, all the eigenstates are unbound, so the eigenvalue E is a complex number,

$$E = \mathcal{E} - \frac{i}{2}\Gamma. \quad (9)$$

Its real part $\mathcal{E}$ gives the energy of the state, while its imaginary part defines the ionization rate Γ . Equations (7) and (8) are solved using the slow-variable discretization (SVD) method [3] in combination with the R -matrix propagation technique [4]. The numerical procedure is

based on discrete variable representations (DVR) [5] constructed from appropriate classical orthogonal polynomials compatible with the boundary conditions [6]. We refer the reader to [1] for more mathematical and numerical details.

In the general cases, for an arbitrary molecular potential $V(\mathbf{r})$, the variables (ξ, η) and φ cannot be separated. In order to solve the molecular problem we have to take into account a coupling between the different m -components. To this end, we rewrite Eq. (1) in the form

$$\left[\frac{\partial}{\partial \eta} \eta \frac{\partial}{\partial \eta} - \frac{1}{4\eta} \frac{\partial^2}{\partial \varphi^2} + \mathcal{B}(\eta) + \frac{E\eta}{2} + \frac{F\eta^2}{4} \right] \psi(\xi, \eta, \varphi) = 0, \quad (10)$$

where

$$\mathcal{B}(\eta) = \frac{\partial}{\partial \xi} \xi \frac{\partial}{\partial \xi} - \frac{1}{4\xi} \frac{\partial^2}{\partial \varphi^2} - \frac{\xi + \eta}{2} V(\xi, \eta, \varphi) + \frac{E\xi}{2} - \frac{F\xi^2}{4}. \quad (11)$$

3. Numerical procedure

In the atomic case [1], the eigenfunctions of the adiabatic Hamiltonian (8) are obtained using the DVR basis constructed from the generalized Laguerre polynomials $L_n^{(|m|)}(s\xi)$ [6], where the scaling factor s defines the extent of the basis functions in ξ . This approach enables one to exactly incorporate the regularity boundary con-

dition $\psi(\xi, \eta)|_{\xi \rightarrow 0} \propto \xi^{|m|/2}$ into the formulation. In the molecular case, however, the solution $\psi(\xi, \eta, \varphi)$ contains integer as well as half-integer powers of ξ for $\xi \rightarrow 0$, which cannot be represented by a single Laguerre-DVR basis with a fixed m . To resolve this difficulty, we introduce a new variable,

$$\xi = \zeta^2. \quad (12)$$

Then

$$\mathcal{B}(\eta) = \frac{1}{4\zeta} \left[\frac{\partial}{\partial \zeta} \zeta \frac{\partial}{\partial \zeta} + \left(\frac{1}{\zeta} + \frac{\zeta}{\eta} \right) \frac{\partial^2}{\partial \varphi^2} - 2\zeta(\zeta^2 + \eta)V(\xi, \eta, \varphi) + 2E\zeta^3 - F\zeta^5 \right]. \quad (13)$$

This transformation allows us to use a single Laguerre-DVR basis with $m = 0$ for the expansion in ζ . The eigenfunctions of $\mathcal{B}(\eta)$ are constructed by using the direct product of the Laguerre-DVR basis in ζ and a sinusoidal basis with periodic boundary conditions in φ .

In the R -matrix theory [7], the space is divided into inner and outer regions. The inner region $0 \leq \eta \leq \eta_c$ is divided into N_{sec} sectors, $0 = \eta_0 < \eta_1 < \dots < \eta_{N_{\text{sec}}} = \eta_c$. In each sector, we construct the R -matrix basis $\bar{\psi}_n(\xi, \eta, \varphi)$ defined by

$$\left[\frac{\partial}{\partial \eta} \eta \frac{\partial}{\partial \eta} - \mathcal{L} - \frac{1}{4\eta} \frac{\partial^2}{\partial \varphi^2} + \mathcal{B}(\eta) + \frac{\bar{E}_n \eta}{2} + \frac{F\eta^2}{4} \right] \bar{\psi}_n(\xi, \eta, \varphi) = 0, \quad (14)$$

where $\mathcal{L}$ is the Bloch operator,

$$\mathcal{L} = \eta[\delta(\eta - \eta_+) - \delta(\eta - \eta_-)] \frac{\partial}{\partial \eta}. \quad (15)$$

In the atomic problem [1], in order to incorporate the regularity boundary condition $\psi(\xi, \eta)|_{\eta \rightarrow 0} \propto \eta^{|m|/2}$, in the first sector Eq. (14) was solved using the DVR ba-

sis constructed from the Jacobi polynomials $P_n^{(0, |m|)}(x)$ [6]; in further sectors Legendre polynomials were used. However, in the molecular case the solution contains integer as well as half-integer powers of η for $\eta \rightarrow 0$, similarly to the ξ variable. To allow the representation of the solution by

a single Legendre-DVR basis, we need to implement another change of variables in the first sector,

$$\eta = \chi^2. \quad (16)$$

In the outer region, $\eta > \eta_c$, the molecular potential is substituted by a purely Coulomb tail $-Z_{as}/r$. Then the problem becomes exactly separable in parabolic coordinates and reduces to solving uncoupled equations in η [1]. Here the outgoing-wave boundary condition is applied. The R -matrix is propagated from $\eta = 0$ outward, and from $\eta = \eta_c$ inward. By matching the solutions at the right boundary of the first sector, we obtain the Siegert eigenvalue E and eigenfunctions.

Some preliminary results for an H_2^+ molecule aligned along the electric field are presented in the figures. The energy $\mathcal{E}$ and ionization width Γ of the lowest sigma state as function of the field F are shown in Fig. 1. The transverse momentum distribution of the ionized electrons for $F = 0.5$ a.u. is shown in Fig. 2.

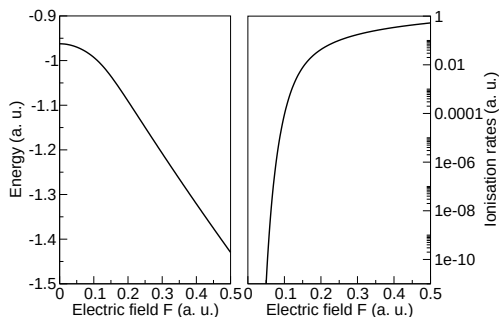


Fig. 1. Energy and Ionisation rates as a function of electric field F .

4. Conclusion

In this report, we have briefly presented the development necessary to calculate the molecular Siegert states in a static electric field. Starting from an atomic problem enabling the calculation of Siegert states in the single active electron approximation, we have extended the program by reducing the use of the symmetries and including m -coupling to solve the molecular problem. This new method enables one to obtain the eigenvalue

and eigenfunction for a particular Siegert state as a functions of the electric field F for molecules modeled by one-electron potentials.

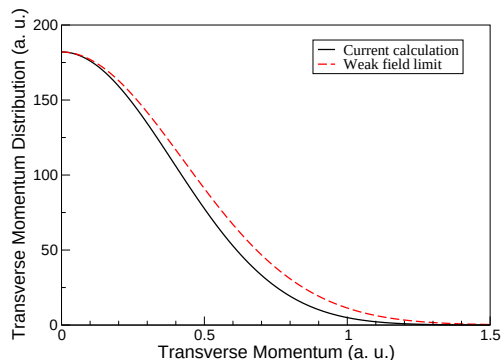


Fig. 2. Transverse momentum distribution as a function of transverse momentum. The current calculation is compared to the weak field limit Gaussian defined by $\exp(-\frac{K_{\perp}^2 \sqrt{2|E_0|}}{F})$ with $F = 0.5$ a.u., the electric field, $|E_0|$ the ionisation potential without laser field and $K_{\perp}$ the transverse momentum.

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Finite Size Effects in High-Intensity QED

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Introduction

Within the next few years lasers are expected to reach unprecedented extremes in power (>10 PW), intensity ($>10^{23}$ W/cm²) and, accordingly, field strength ($>10^{15}$ V/m). In these regimes, equivalent to concentrating the total solar radiation on a pinhead, both relativistic and quantum effects need to be taken into account. The appropriate theoretical tool is a relativistic quantum field theory, namely strong-field, or high-intensity, quantum electrodynamics (QED). This may also be understood from the fact that one is approaching the QED critical electric field strength^[1,2],

$$E_S = \frac{m^2 c^3}{e \hbar} = 1.3 \times 10^{18} \text{ W/m}, \quad (1)$$

which depends on the fundamental constants of both relativity, c , and quantum mechanics, $\hbar$. Beyond this Sauter-Schwinger limit any laser turns into a pair creation machine^[3], as the energy gain of an electron over a Compton wavelength equals its rest mass.

Like any quantum field theory, high-intensity QED is characterised by its particle content. Ordinary photons (Fig. 1, wavy line) serve as the usual virtual exchange bosons or as real particles probing the external field given by the laser.



Figure 1: Wavy line: Probe photon; dashed line: laser photon.

The associated laser photons (Fig.1, dashed line), on the other hand, need in general not be explicitly shown. They rather play an ‘auxiliary’ role by ‘dressing’ the electrons whereupon these become effective particles, represented by the double line in Fig. 2. Formally, they are given by the Volkov solution^[4] of the Dirac equation in a plane wave field, $F^{\mu\nu} = F^{\mu\nu}(k.x)$, where k is the wave four-vector and $k.x = \omega t - k z$ the invariant phase (assuming propagation along the z -axis).

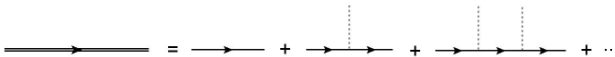


Figure 2: ‘Dressed’ Volkov electron.

The strength of the laser field is measured by the dimensionless laser amplitude,

$$a_0 \equiv \frac{e E \lambda_L}{m c^2}, \quad (2)$$

the energy gain of a probe electron in the ambient electric field E across a laser wavelength $\lambda_L = 2\pi c/\omega$, divided by the electron rest energy¹. Defining a dimensionless electric field and frequency via

$$\epsilon \equiv E/E_S, \quad \nu \equiv \hbar\omega/mc^2, \quad (3)$$

one may alternatively write $a_0 = \epsilon/\nu$. For a laser one has $\epsilon < 1$, an upper bound imposed by the Sauter-Schwinger limit, which implies that $a_0 < 1/\nu < 10^6$ for an optical laser. For the Vulcan-10PW and Extreme Light Infrastructure (ELI) facilities one expects $a_0 \approx 10^2$ and $a_0 \approx 10^3$, respectively. The parameter a_0 also serves as a characteristic of the Volkov electrons of Fig. 2. They acquire a quasi-momentum given by²

$$q_\mu = p_\mu + \frac{a_0^2 m^2}{2k \cdot p} k_\mu, \quad (4)$$

with a longitudinal addition, proportional to a_0^2 , to the asymptotic particle momentum p ^[6,7]. Upon squaring one recovers

$$q^2 = m^2(1 + a_0^2) \equiv m_*^2, \quad (5)$$

the celebrated intensity dependent mass shift of an electron in a laser background first discovered by Sengupta^[6] and thoroughly discussed by Kibble^[8]. The mass shift (5) may be understood in purely classical terms. Solving the Lorentz equation for an electron in an infinite plane wave (IPW) one finds the momentum as a function of proper time, $p = p(\tau)$. The quasi-momentum (4) is then the proper time average of $p(\tau)$, taken over a laser period, the asymptotic momentum p serving as an initial condition.

With the basic intensity, or a_0 , dependence of the Volkov electrons identified we may use their graphic representation (Fig. 2) together with a probe photon line from Fig. 1 to build the ‘fundamental’ vertex of laser-induced strong-field QED. Its Feynman graph is depicted in two different ways in Fig. 3.

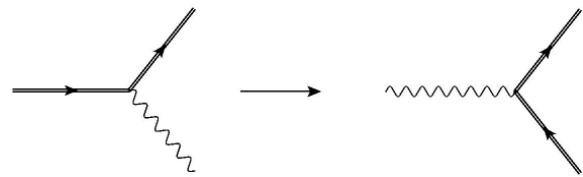


Figure 3: Two versions of the basic strong-field QED interaction, viewed as transition amplitudes, represent nonlinear Compton scattering (left) and stimulated pair production (right). They are related by crossing symmetry.

Several comments are in order. The vertex is fundamental in terms of Volkov electrons but, once expanded via Fig. 2 it becomes an infinite series in terms of diagrams with more and more laser photons. The Volkov representation thus is a rather economic resummation of many-particle tree diagrams and as such is *nonperturbative* in the coupling e . Viewing the graphs of Fig. 3 as S -matrix elements (transition amplitudes) rather than vertices the two diagrams acquire different meanings. The left-hand graph describes nonlinear Compton scattering (NLC) or, equivalently, the emission of a photon by an electron in an intense wave^[9,10]. (See [11] for a recent reanalysis and update.). Expanding the Volkov lines according to Fig. 2 yields a sum over all processes of the form

¹ This can be written in a manifestly Lorentz and gauge invariant way^[5].

² We adopt natural units, $\hbar=c=1$, in what follows.

$$e + n_1 \gamma_L \rightarrow e' + \gamma', \quad (\text{NLC}) \quad (6)$$

labelled by n_1 , the number of laser photons involved. The right-hand graph of Fig.3 represents stimulated pair production (sPP) with the ‘stimulating’ photon, γ' , apparently decaying into a Volkov electron-positron pair. Again, this may be viewed as summing all processes,

$$\gamma' + n_2 \gamma_L \rightarrow e^+ + e^-, \quad (\text{sPP}) \quad (7)$$

over n_2 . Whenever the laser photon numbers $n_1, n_2 > 1$, the process in question is called nonlinear. The linear processes ($n_1 = n_2 = 1$) are standard Compton scattering^[12] and Breit-Wheeler pair production^[13]. In the limit of vanishing external field (formally obtained by setting $n_1 = n_2 = 0$), i.e. for bare rather than dressed electrons, both scattering processes are forbidden by energy-momentum conservation.

The two strong-field processes (6) and (7) were simultaneously observed in the famous experiment SLAC E-144^[14,15,16] with values of n_1 and n_2 up to five. In particular, about 10^2 positron events were recorded, realising the ‘creation of matter from light’ for the first time. The experimental results have been claimed to be consistent with the theory as developed in [9,10]. It should, however, be emphasised that this theory was explicitly worked out only for plane waves of infinite temporal and spatial extent. In general, however, such an IPW does not represent a realistic laser beam which typically is pulsed (hence of finite time duration) and also localised in space, both longitudinally and transversally, as measured by Rayleigh length, z_R and waist, w_0 . It thus seems appropriate to address possible modifications of the theory and its predictions caused by finite spatio-temporal size effects.

Finite Size Effects in Nonlinear Compton Scattering

As already noted, strong-field QED in laser backgrounds relies heavily on the existence of the Volkov solution in a plane wave field, $F^{\mu\nu} = F^{\mu\nu}(\varphi)$, $\varphi = k \cdot x$. The wave in question, however, may be an *arbitrary* function of φ . Hence it need not be of infinite extent in φ as for an IPW, but may be a finite wave train^[17] or pulsed with a smooth envelope. We will generically refer to such configurations as pulsed plane waves (PPWs). A nice example of the latter has recently been discussed by Mackenroth et al.^[18] with a pulse profile which we slightly generalise to

$$f(\phi) = \begin{cases} \sin^K(\phi/2N) \sin(\phi + \phi_0), & 0 \leq \phi \leq 2\pi N \\ 0, & \text{otherwise} \end{cases} \quad (8)$$

allowing for an additional ‘carrier phase’ ϕ_0 between the oscillatory sine part and the $\sin^K$ envelope³. The integer N counts the number of cycles within the pulse, see Fig. 4.

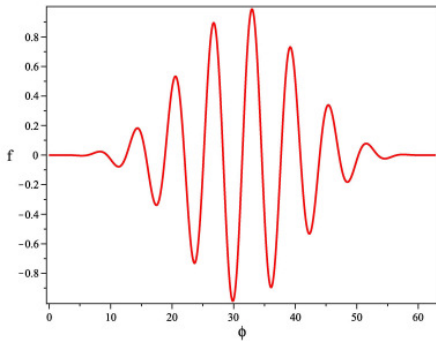


Figure 4: Pulse profile (8) for $K=4, N=10$ and $\phi_0=0$.

³ Mackenroth et al.^[18] used $K=4$ and were particularly interested in the carrier phase and its consequences. For our purposes, though, it is sufficient to set $\phi_0=0$.

Soon after the mass shift had been (re)discovered in the sixties there was a discussion on whether it is really observable in view of the somewhat unphysical nature of an IPW. Kibble^[8] has convincingly argued (using an exponentially damped wave) that all that is required are pulses that are ‘sufficiently’ long. Quantitatively, this translates into having a small width $\Delta\omega$ in laser frequency, $\Delta\omega \ll \omega$, which just corresponded to the state of the art for lasers in the sixties⁴. In terms of the invariant phase this may be reformulated as

$$\Delta\phi/2\pi \sim N \gg 1, \quad (9)$$

where $\Delta\phi$ is the width in ϕ , taken to be $2\pi N$ for the pulse (8). So we expect an IPW to be a reasonable model in the large- N limit, i.e. when the laser pulse contains many cycles. The SLAC E-144 100 TW laser pulse had a duration of 2 ps corresponding to $N \approx 10^3$ which should be close to the IPW regime. For current ultra-short pulses of, say, 10 fs, however, one clearly enters the few-cycle regime, $1 < N < 10$, where Kibble's criterion (9) ceases to be valid. This suggests that the idea of an effective mass arising as an averaging effect due to the electron quiver motion may become questionable. As the Volkov solution remains valid also for pulses it is possible to test the significance of mass shift effects quantitatively. While the IPW S -matrix elements^[9,10] can be extended to the PPW case, there is an alternative route available for NLC in the classical (Thomson) limit ($v \ll 1$). This is to calculate the classical Larmor radiation (or bremsstrahlung) of an electron accelerated by the wave. This has been done in [19] where the validity of the classical limit was also checked explicitly. The complete IPW spectrum of the scattered photons is shown in Fig. 5.

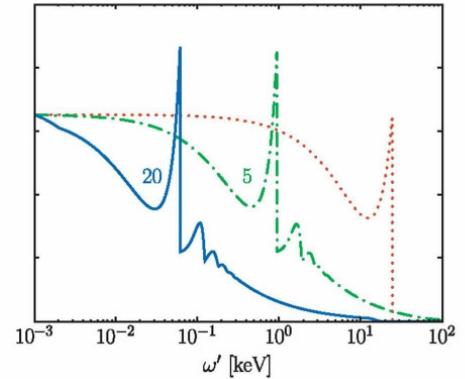


Figure 5: Total photon emission spectrum (in arbitrary units) for NLC, summed over all harmonics, as a function of scattered frequency, $\omega' = mv'$. Parameter values: $a_0 = 5$ and $a_0 = 20$ as indicated, $\gamma = 80$. The dotted curve corresponds to $a_0 \rightarrow 0$, i.e. standard (‘linear’) Compton scattering. (From [19].)

The following generic intensity effects may be noted: (i) the kinematic $n_1 = 1$ Compton edge (represented by the principal peak in each of the spectra) is red-shifted, when a_0 is nonzero, to the value

$$\nu'_1(a_0) = \frac{\nu e^{2\zeta}}{1 + a_0^2 + 2\nu e\zeta} \simeq \frac{4\gamma^2\nu}{1 + a_0^2}. \quad (10)$$

Here, ζ denotes rapidity i.e. $e^\zeta = \gamma(1+\beta)$, with γ and β the standard Lorentz factors for the incoming electron. The last identity in (10) is valid when $1 \ll \gamma \ll 1/4\nu$.

(ii) There are smaller peaks beyond the $n_1 = 1$ edge ($v' > \nu'_1$) corresponding to higher harmonics, $n_1 > 1$. The associated spectral ranges are given by $n_1\nu < v' < \nu'_{n1}$, where ν'_{n1} is given by (10) with ν replaced by $n_1\nu$.

⁴ Recall that an IPW corresponds to a delta function, $\delta(\omega)$, hence zero width.

Higher harmonics, i.e. scattered photons with frequencies $\nu' > \nu'_1$, have been observed in the SLAC E-144 experiment together with a ‘shoulder’ corresponding to the $n_1 = 2$ peak (see Fig. 39 in [16]). This was a high energy experiment with 46.6 GeV electrons ($\gamma \approx 0.4$), the Compton edges accordingly of similar magnitudes, $m\nu'_1 = 29$ GeV, $m\nu'_2 = 36$ GeV.

To analyse the effects of finite pulse duration let us concentrate on the first harmonic, $n_1 = 1$. For an IPW this is plotted in Fig. 6 as a function of scattered frequency. The latter has been rescaled by $\nu'_0 = \nu e^{2\zeta}$ such that the linear and nonlinear edges are located at 1 and $1/(1 + a_0^2)$, respectively.

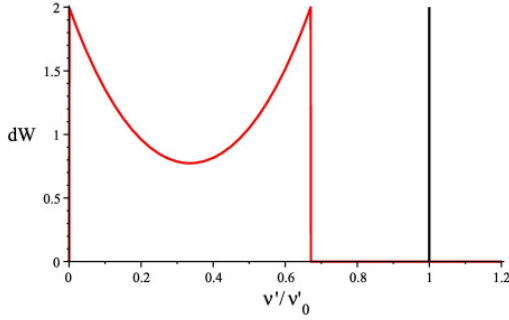


Figure 6: IPW photon emission spectrum (in arbitrary units) corresponding to the first harmonic ($n_1 = 1$), for $a_0 = 0.7$, as a function of rescaled frequency. Note the spectral gap between nonlinear and linear Compton edges (red and black vertical lines, respectively).

The first harmonic signal for an IPW has a sharp edge and there is no signal beyond. Thus, there is a characteristic spectral gap between the nonlinear and linear kinematic edges. This, however, changes significantly for PPWs.

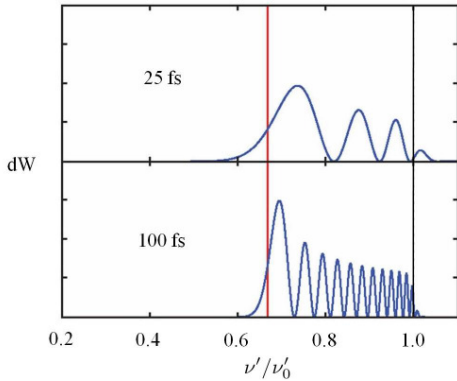


Figure 7: PPW photon emission spectra (in arbitrary units) corresponding to the first harmonic ($n_1 = 1$), for $a_0 = 0.7$, as a function of rescaled frequency. Upper panel: Pulse duration 25 fs. Lower panel: Pulse duration 100 fs. (From [19].)

Fig. 7 shows the first harmonic signal in the same rescaled frequency range for pulse durations of 25 fs and 100 fs. One clearly observes a strong ‘leakage’ of the signal into what was previously the spectral gap. The number of oscillations seems to be linearly increasing with pulse duration, from $N = 3$ to $N = 12$. This suggests that the IPW limit is obtained by having destructive interference of rapid oscillations, an issue which clearly deserves further study. In the very same frequency domain, the signal will be further distorted by the presence of higher harmonics, $n_1 > 1$, recall the discussion of Fig. 5. Thus, for short pulses, it should be rather difficult to observe the red-shift of the Compton edge.

Let us conclude this section with a few words on the effect of the transverse beam profile. There are two complementary cases corresponding to a strong and weak laser focus, respectively (see Fig. 8). The spatial scale to be compared to in both cases is

the electron beam radius, r_b , which, typically, will be of the order of 10 microns.

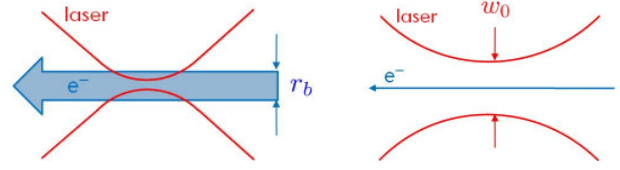


Figure 8: Schematic sketch of laser beams strongly and weakly focused in comparison to the electron beam. Left panel: strong focus, $w_0 < r_b$. Right panel: weak focus, $w_0 \gg r_b$.

For a tight focus, (waist size $w_0 < r_b$, usually required for large $a_0 \gg 1$) the electrons are rather sensitive to the finite transverse extension of the laser beam, and finite size effects will be important. On the other hand, a weakly focussed laser ($w_0 \gg r_b$) will appear approximately as a plane wave (of finite longitudinal extent) to the electrons. In this case, finite size effects should be minor, and strong-field QED based on the Volkov solution should be a reasonable approximation. Indeed, one finds that the strongly focussed case generically leads to finite size dominated, broad and unstructured spectra, while for a sufficiently weak focus spectral substructure remains identifiable^[19].

The SLAC experiment had a laser waist $w_0 \approx 5$ microns and an electron beam radius $r_b \approx 60$ microns $> w_0$, corresponding to the scenario of Fig. 8, left panel. This suggests that effects due to the finite transverse extension of the laser beam should have been significant. In the analysis of the experiment this was dealt with by the ‘adiabatic approximation’ that the IPW theory is valid within small space-time cells covering the laser focus. In each such cell the instantaneous and local value of a_0 (i.e. photon density) as well as the electron density were assumed to be constant (cf. App. A.5 of [16]). It seems difficult to assess the validity of this assumption.

Finite Size Effects in Stimulated Pair Production

As stated in the introduction, the sPP process (7) has also been observed at SLAC by (i) detecting about 10^2 positron signals and (ii) finding a nonlinear dependence on the number of laser photons^[15,16]. Note that, unlike NLC (6) sPP is a threshold process. For an IPW, energy-momentum conservation involves the electron and positron quasi-momenta, $k' + n_2 k = q + q'$, which makes the threshold dependent on the *effective* mass (5), requiring that the usual Mandelstam variable $s > 4m_*^2$ or, in terms of dimensionless parameters and assuming a head on collision,

$$n_2 \geq \frac{1 + a_0^2}{\nu \nu'} . \quad (11)$$

Clearly, the minimum n_2 increases with a_0 and decreases with the energy, ν' , of the ‘stimulating’ photon which, in the SLAC experiment, was obtained by NLC back-scattering resulting in $m\nu' \approx 29$ GeV for $n_1 = 1$ or $m\nu' \approx 36$ GeV for $n_1 = 2$. In the parameter range $0.2 < a_0 < 0.4$ SLAC E-144 determined a positron production rate R which was well fitted by an intensity dependence according to the power law

$$R \sim a_0^{2n} , \quad n = 5.1 \pm 0.2 (\text{stat})_{-0.8}^{+0.5} (\text{syst}) , \quad (12)$$

clearly indicating an exponent $n = 5$ (cf. Fig. 44 of [16]). This has been interpreted as the requirement of having 5 laser photons participating for the SLAC a_0 range just given. Note, however, that the processes (6) and (7) were both happening repeatedly and simultaneously in the space-time volume (given by the overlap of laser and electron beams) and thus were impossible to disentangle experimentally, and so the n from

(12) should be a linear combination, $n = n_1 + n_2$, stemming from *both* processes. With $n_1 = 1$ producing a 29.1 GeV photon, the situation here is a little subtle: according to (11), the required number of *additional* photons required to produce a pair changes from $n_2 = 4$ to $n_2 = 5$ at $a_0 \approx 0.22$. Hence, the SLAC results seem to exhibit some ‘sub-threshold’ production of pairs. Note also that for pairs produced when $a_0 < 0.22$, both (11) and the ‘linear’ ($a_0 = 0$) threshold predict $n_2 = 4$ for the photon number. Thus it seems that $n_1 = 1$ and $n_2 = 4$ cannot give an unambiguous identification of effects caused by the mass shift. The upshot then is the following. While SLAC E-144 has clearly identified *nonlinear* behaviour ($n_1, n_2 > 1$) the a_0 values may have been too small for a definite identification of intensity effects in sPP such as the shifted threshold. Hence, Sengupta’s mass shift^[6,8] remains unobserved.

In light of the above, we now turn to consider the effects of finite pulse duration^[20]. Again, also for sPP, strong-field QED based on Volkov solutions remains valid for pulses in $\varphi = k \cdot x$. The main consequences are summarised in Fig. 9 showing a triple differential sPP rate, d^3W , assuming the pulse (8) with $K = 0$, i.e. a finite wave train with no envelope.

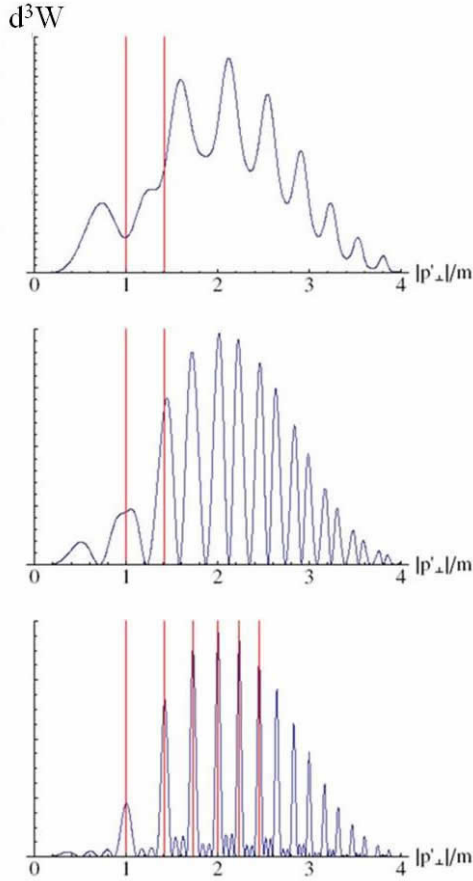


Figure 9: Triple differential sPP rate (arbitrary units) as a function of transverse positron momentum. Circular polarisation, $a_0 = 2$, $mv' = 250$ GeV, $N = 1, 2$ and 4 cycles per pulse (descending), i.e. pulse durations of 4 fs, 8 fs and 16 fs, respectively. Vertical lines show the IPW limit (a delta comb).

Most interestingly, one observes another ‘leakage’ phenomenon very much analogous to NLC (cf. Figs. 6 and 7). For an IPW the rate is given by delta comb resonances. These are shown in Fig. 9 as vertical red lines which begin, for our parameters, at a threshold value of $lp'_\perp/m = 1$ where $p'_\perp$ denotes the transverse positron momentum. For a PPW, however, the signal ‘leaks’ into the transverse momentum regime below, in other words, there is *sub-threshold* behaviour, i.e. for centre-of-mass energies below $2m_*$.

This behaviour is most pronounced when the number N of cycles per pulse defined in (9) is small (first panel of Fig. 9). In the large- N limit the IPP features are recovered (last panel of Fig. 9). Thus, an unambiguous observation of the mass shift in sPP (in terms of a blue-shifted sPP threshold) will require $a_0 \gg 1$, hence short pulses, and, accordingly, a significant control of finite pulse duration effects.

We remark that the rapid oscillations appearing in both Fig. 7 and Fig. 9 are a typical signal of finite (temporal) pulse duration: they are also observed in the spectra of pairs produced in the focus of colliding lasers, modelled by oscillating electric fields^[21,22].

Conclusions

We have discussed two basic processes of high-intensity QED, nonlinear Compton scattering (NLC) and stimulated pair production (sPP). These processes are related by crossing symmetry and hence show some similar features. If the laser is modelled as an infinite plane wave, intensity effects manifest themselves as clear and unambiguous signals, ultimately caused by the intensity dependent mass shift, $m^2 \rightarrow m^2(1 + a_0^2) = m_*^2$. For both processes, these signals are given by spectral shifts in the allowed kinematic regions compared to the ‘linear’ ($a_0 = 0$) processes. For NLC one finds a red-shift of the upper Compton edge in the spectrum of emitted photons. For sPP, on the other hand, there is a blue-shift of the sPP threshold directly proportional to m_* . Both effects increase with intensity, hence with laser amplitude, a_0 , which seems to favour having large a_0 . However, the standard way of boosting a_0 is temporal pulse compression. On the theoretical side this implies trading the infinite plane wave model for a more realistic finite pulse model which is reasonably straightforward. In such models, pulse duration is conveniently measured by the number N of laser cycles per pulse. For small $N = 1 \dots 10$, the spectral shifts characterising the infinite plane wave model get washed out, the more so the smaller N . It thus seems that an unambiguous identification of intensity effects on particle spectra will require a delicate balance between having sufficiently large $a_0 \gg 1$ (high compression) and sufficiently long pulses (low compression). As these requirements are somewhat in opposition of each other, a substantial amount of fine-tuning will be necessary. Nevertheless, it is possible to identify parameters which help to expose various intensity effects^[19], and we are optimistic that their observation will remain feasible.

Acknowledgements

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Multi-electron dynamics on the femtosecond time-scale

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Introduction

The recent emergence of attosecond light sources promises to provide deeper insight into ultrafast *correlated* dynamics between electrons in complex matter [1]. In order to accurately describe such dynamics, theoretical methods which go beyond a single-active-electron (SAE) description of the laser-atom interaction are required. One method that offers such a description is the time-dependent *R*-matrix (TDRM) method, recently developed at Queen's University Belfast [2]. In the first part of this report, we use the TDRM method to investigate ultrafast correlated dynamics between two 2p electrons inside C^+ . We show that, by selecting laser pulse parameters carefully it is possible to choose the nature of the correlated dynamics between these two 2p electrons. In the second part of the report we investigate how the choice of dipole gauge can affect the convergence of TDRM calculations. Our findings help to verify that for the interaction of the laser field with a multi-electron atom, the laser field near the nucleus is in practice best described using the length form of the dipole operator.

Time-dependent *R*-matrix theory

In TDRM theory, the time-dependent Schrödinger equation is solved by dividing the position space of the atomic electrons into an inner and outer region with the boundary of the inner region at radius $r = a_0$ and the boundary of the outer region at radius $r = a_p$ (see figure 1) [3]. In the inner region, electron exchange and correlation effects between the ejected electron and the remaining electrons are important and atom-laser Hamiltonian matrix elements are calculated explicitly. In the outer region only one electron is present, which besides experiencing the laser field directly, is aware of the remainder of the atomic system only via long-range multi-pole interactions.

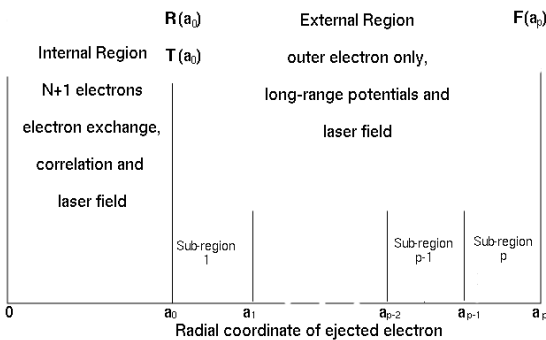


Figure 1. Division of the position space of the atomic electrons in TDRM theory.

In the inner region, an *R*-matrix basis expansion of the wave function is adopted. Using a linear solver at each time step, we can calculate the *R*-matrix, $\mathbf{R}$, on the boundary, $r = a_0$, of this region and also calculate an inhomogeneous *T*-vector, $\mathbf{T}$, both as defined in [2].

In the outer region $a_0 \leq r \leq a_p$, a set of coupled differential equations describing the motion of the ejected electron in the presence of the light field needs to be solved at each time step.

We achieve this by sub-dividing this region into p sub-regions and propagating the *R*-matrix and *T*-vector across them from $r = a_0$ to $r = a_p$ as shown in figure 1. The *R*-matrix and *T*-vector at $r = a_p$ can be used to propagate the wave function ($\mathbf{F}$ in figure 1) backwards across the p sub-regions with the assumption $\mathbf{F}(a_p) = 0$. This propagated wave function then provides the starting point for the calculation at the next time step.

Influencing correlated two-electron dynamics inside C^+

We propose a pump-probe scheme in which we are able to influence the nature of the correlated dynamics occurring between two electrons [4]. Our scheme is similar to a pump-probe scheme used in a previous study of collective electron dynamics [5]. The basic features of the scheme are presented in figure 2.

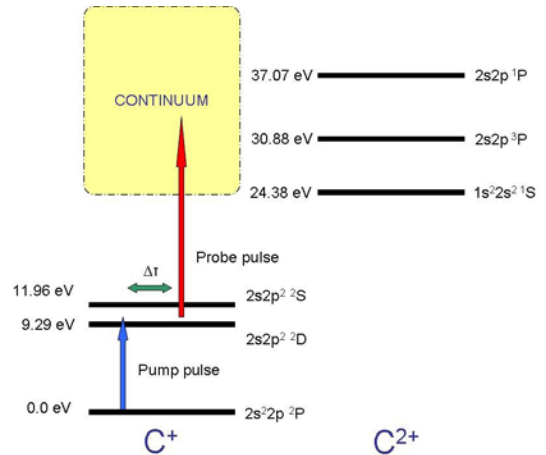


Figure 2. Pump-probe scheme. The pump pulse has a photon energy, $\omega_1 = 10.0$ eV in order to be near resonant with the 2S and 2D states, but with different detunings. The time-delayed probe pulse has a photon energy, $\omega_2 = 21.8$ eV to transfer population above the $C^{2+} 2s2p^3 \ ^3P$ ionization threshold.

We consider C^+ in its ground state with total magnetic quantum number $M=0$. The ion is excited by an XUV pulse linearly polarized in the z direction into a superposition of the excited $2s2p^2 \ ^2S$ and 2D states. The repulsion between the two 2p electrons results in interference between the two excited LS states, which occurs on a time scale of 1-2 femtoseconds. By subsequently ionizing C^+ with a time delayed ultrashort XUV pulse and analyzing the properties of the ejected electron we are able to probe the interference and investigate the role of electron-electron interactions in the transition from ultrashort-pulse excitation to long-pulse excitation.

Results

To describe the correlated dynamics of the 2p electrons in terms of the dynamics of individual electrons, we transform from the LS-coupled basis to the uncoupled $|2p_{m1} 2p_{m2}\rangle$ basis, in which the roles of magnetic sub-states becomes more transparent.

Figure 3 shows the 2-dimensional (2D) momentum distributions of the ejected electron obtained for three separate pump pulse durations and for three delays of the probe pulse.

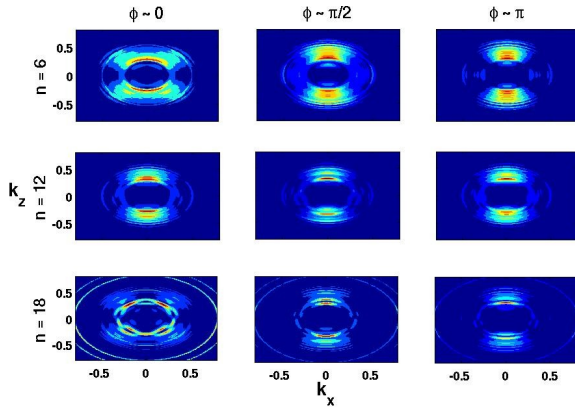


Figure 3. 2D momentum distributions associated with the residual C^{2+} ions left in the $2s2p\ ^3P$ state in the k_x, k_y plane for the ejection of an electron from the pumped C^+ ion for pump pulse durations of $n = 6, 12$ and 18 cycles. ϕ corresponds to the phase of the breathing motion.

The first time delay ($\phi \sim 0$) between the pulses corresponds to a minimum of the $|2p_0\ 2p_0\rangle$ population. The third delay ($\phi \sim \pi$) corresponds to a maximum $|2p_0\ 2p_0\rangle$ population and the second delay ($\phi \sim \pi/2$) corresponds to the midpoint between these two delays. Figure 3 clearly shows the dominant emission of $|m| = 1$ electrons for the $n = 6$ and $n = 18$ cycle pump pulses for delays at which the $|2p_0\ 2p_0\rangle$ is at a minimum. At the third time delay, the probe pulse interacts with the wave-packet when the $|2p_0\ 2p_0\rangle$ is at a maximum. The dominance of the $m = 0$ electron emission along the laser polarization axis is clearly evident at this delay. This behavior indicates that the wave-packet is undergoing a breathing motion in which it oscillates between two distinct angular distributions. The wave-packet will oscillate between these distributions after the end of the pulse with a period of 1.5 fs, determined by the energy separation between the 2S and 2D states. For a wave-packet prepared by the $n = 12$ cycle pump pulse, however, it is more difficult to find evidence of correlated electron dynamics. The dominance of population in the 2D state means that the breathing motion between the two angular distributions only forms a small part of the excitation into the $2s2p^2$ configuration.

Figure 3 shows that by carefully choosing the duration of the excitation pulse, it is possible to select the nature of the excitation of the $2p^2$ configuration. For certain excitation pulse durations, the excitation will be dominated by a single LS coupled state, but for other pulse durations, the excitation will be dominated by a breathing motion between uncoupled states.

Choice of dipole gauge in the TDRM method

One of the main challenges in theoretical atomic physics is the description of multi-electron dynamics in intense laser fields. A full description of multi-electron dynamics is computationally demanding and, consequently, approximations must be made. In the TDRM method we use a restricted basis set to describe the system. As a result of this approximation, the choice of dipole operator gauge used to describe the laser-atom interaction has a significant effect on the outcomes.

We investigate the optimum choice of gauge for use with the TDRM method by considering a test case of multi-photon ionization of He using both the dipole length gauge and the dipole velocity gauge [6]. The basis sets we consider range from using only the 1s orbital to describe He^+ , to including the 1s, 2s, 2p, 3s, 3p and 3d orbitals (and pseudo-orbitals) in the basis set for He^+ .

Results

Figure 4 shows the ionization probabilities as a function of time for a laser with a photon energy $\omega = 6.53$ eV for both the length gauge (solid lines) and velocity gauge (dashed lines) when

using two different basis sets. The length gauge provides a more consistent result, with the velocity gauge showing a large discrepancy between each basis set. Ultimately both gauges converge to the same result as additional orbitals and pseudo-orbitals are added to the basis set.

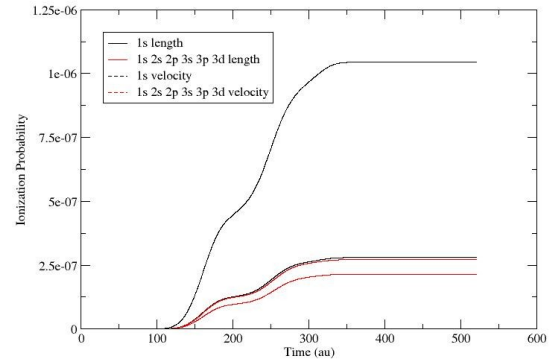


Figure 4. Helium ionization probabilities using the length and velocity gauges for a laser pulse with photon energy 6.53 eV and peak intensity 2×10^{13} W cm $^{-2}$.

As the length gauge is found to have a higher consistency with respect to the number of orbitals in the basis set, we consider the length gauge to be the preferred choice of gauge. Analysis of multiphoton matrix elements suggests this is a result of a higher sensitivity to high energy processes in the velocity gauge.

Conclusion

In the first part of this report we have used the recently developed TDRM method to propose a technique that may provide the possibility of influencing the field-free evolution of multi-electron wave-packets. In the near future, we intend to investigate more elaborate laser pulse schemes which will allow more control over the population of the excited states during the pumping stage. We also intend to extend this study to more complex configurations. Such studies may lead to further insight into the fundamental connection between atomic structure and correlated multi-electron dynamics.

In the second part of the report we investigated how the choice of dipole gauge can affect TDRM calculations and have shown that for a laser field interacting with a multi-electron atom close to nucleus, the length form of the dipole operator appears to be a better choice of gauge. This is particularly the case for the finite basis set calculations employed in the TDRM inner region.

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Hydrodynamics of the Yukawa One Component Plasma

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Introduction

The hydrodynamic description is a very common starting point for investigating a wide range of basic and complex physical phenomena in fluids. In this macroscopic context, the behaviour of the fluid is governed by the equations of fluid mechanics [1] (e.g. the Navier-Stokes equations). These equations give a wide range of physical predictions (hydrodynamic modes, etc.) that are frequently tested and verified experimentally (e.g. [2]).

From a microscopic standpoint, usually the hydrodynamic regime or *hydrodynamic limit* corresponds to wavenumbers k such that $kl_f \ll 1$ with l_f the 'mean free path' and frequencies ω such that $\omega\tau_c \ll 1$ with τ_c the 'mean collision time'. These conditions, which are already rather qualitative for a system governed by uncorrelated binary collisions, become even more indeterminate when many-body correlations are present because the concepts of 'mean free path' and 'mean collision time' cease to have a clear physical meaning. Thus, when these many-body correlations are present (as is the case, for example, in a liquid or dense plasma), the rather general question of the domain of validity of the hydrodynamic limit is particularly challenging. The extent to which these correlations are present in the system depends on the thermodynamic parameters - the density and temperature - as well as the strength and range of the (microscopic) interactions. For instance, one can expect that the hydrodynamic description only applies at lengthscales that are greater than the range r_c of the potential (i.e. $kr_c < 1$) - in other words, that the domain of validity of the hydrodynamic limit will shrink as the range of the potential increases.

In this contribution, we investigate when the hydrodynamic description (rather than a more detailed microscopic description) is applicable for a physical system. To achieve this, we consider the microscopic dynamics of the so-called Yukawa One Component Plasma (YOCP), which has interaction potential $v(r) = q^2 \exp(-r/r_c)/r$, where q^2 is the strength and r_c the range of the potential. Since it is known that this model is fully characterised by two dimensionless parameters [3], we are able to very effectively explore the entire parameter space (density n , temperature T , strength and range of the interaction potential) that influences the applicability of the hydrodynamic description. Specifically, these parameters are the reduced range $r_c^* = r_c/a$, where $a = (4\pi n/3)^{-1/3}$ is (a measure of) the average inter-particle spacing, and the coupling strength $\Gamma = q^2/(ak_b T)$, which itself characterises completely the degree of many body correlations present in the system for a given range. We calculate the complete (equilibrium) microscopic dynamics of this model - specifically, we calculate the so-called dynamical structure factor - and compare these to the dynamics predicted by the hydrodynamic (i.e. macroscopic) description.

Dynamical Structure Factor (DSF)

The space and time dependent density-density correlation function $\langle n(\mathbf{r}, t) n(\mathbf{r}', t') \rangle$ (also known as the Van Hove function) plays a privileged role in statistical physics as a bridge between theory and experiment. Here $n(\mathbf{r}, t)$ is the (microscopic) density of a system of N point particles

$$n(\mathbf{r}, t) = \sum_{i=1}^N \delta(\mathbf{r} - \mathbf{r}_i(t))$$

And $\langle \dots \rangle$ denotes the usual statistical (thermal equilibrium) average, which is given for the canonical ensemble as

$$\langle \dots \rangle = \frac{1}{Z} \int \dots e^{-BH(\mathbf{r}^N, \mathbf{p}^N)} d\mathbf{r}^N d\mathbf{p}^N$$

With Z the canonical partition function. Here B is the inverse temperature $1/k_b T$ and $H(\mathbf{r}^N, \mathbf{p}^N)$ is the Hamiltonian of the system. Working instead with the density fluctuation

$$\delta n(\mathbf{r}, t) = n(\mathbf{r}, t) - \langle n(\mathbf{r}, t) \rangle$$

The intermediate scattering function is defined as

$$F(\mathbf{k}, t) = \frac{1}{2\pi N} \langle \delta n(\mathbf{k}, t) \delta n(-\mathbf{k}, 0) \rangle \quad (1)$$

Where

$$\delta n(\mathbf{k}, t) = n(\mathbf{k}, t) - \langle n(\mathbf{k}, t) \rangle$$

is the density fluctuation in wavevector space, with

$$n(\mathbf{k}, t) = \sum_{i=1}^N e^{-i\mathbf{k} \cdot \mathbf{r}_i(t)}$$

the spatial Fourier transform of the density at time t . Finally then, the Dynamical Structure Factor $S(\mathbf{k}, \omega)$ is the Fourier transform in time of the intermediate scattering function

$$S(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} F(\mathbf{k}, t) e^{i\omega t} dt \quad (2)$$

When the system possesses spherical symmetry (as is the case for the Yukawa model, because the interaction potential depends only on the distance between two particles), the DSF depends only on the magnitude k of the wavevector $\mathbf{k}$, and is thus denoted as $S(k, \omega)$ henceforth. The DSF corresponds experimentally to the spectrum measured from light (e.g. x-ray) or neutron inelastic scattering experiments.

Computing the DSF

Determining the DSF is a case of evaluating the integral given in Eq. (2), which requires calculation of the intermediate scattering function. This cannot generally be done analytically - one notable counterexample being an ideal gas of non-interacting particles. Thus, it is usual to resort to numerical methods for the calculation of the DSF. Of use here is the 'ergodicity assumption' (see e.g. [4]) that allows the statistical average to be computed instead as a time average. Roughly

speaking, the 'ergodicity assumption' amounts to the hypothesis that the dynamical system 'samples' the states in the phase space in time with the same frequency as given by the weighting in the statistical average. Specifically, when applied to the calculation of the intermediate scattering function, one writes mathematically

$$F(k, t) = \frac{1}{2\pi N} \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T \delta n(\mathbf{k}, t+t') \delta n(-\mathbf{k}, t') dt'$$

In this way knowledge of the dynamics of the system (i.e. the trajectories of the particles as given by Newton's equations for the case of a classical system) allows one to calculate a statistical average of a quantity of physical interest (in this case the intermediate scattering function). Therefore, the problem of calculating the DSF reduces to solving Newton's equations for the particle trajectories, which can be done numerically by taking advantage of the long established techniques that generally come under the umbrella term 'Molecular Dynamics' (MD).

Hydrodynamic Description of the DSF

From MD simulations then the 'exact' DSF for the Yukawa model can be calculated to within numerical accuracy. It is this quantity that we wish to compare to the hydrodynamic description, in order to examine when the hydrodynamic description is applicable. This macroscopic description is determined from the linearised hydrodynamic (Navier-Stokes) equations, which are (e.g. [5])

$$\begin{aligned} \frac{\partial}{\partial t} \delta n(\mathbf{r}, t) &= -n \nabla \cdot \mathbf{v}(\mathbf{r}, t) \\ mn \frac{\partial \mathbf{v}(\mathbf{r}, t)}{\partial t} &= -\nabla \delta p(\mathbf{r}, t) + \eta \nabla^2 \mathbf{v}(\mathbf{r}, t) + \left(\zeta + \frac{\eta}{3}\right) \nabla \nabla \cdot \mathbf{v}(\mathbf{r}, t) \\ T \frac{\partial}{\partial t} \delta s(\mathbf{r}, t) &= \frac{\kappa}{n} \nabla^2 \delta T(\mathbf{r}, t) \end{aligned} \quad (3)$$

In Eqs. (3), n and T are the equilibrium number density and temperature respectively, with m the particle mass. The shear and bulk viscosities η and ζ enter into the momentum balance equation with the thermal conductivity κ in the energy balance equation. One can express the entropy and pressure fluctuations ($\delta s(\mathbf{r}, t)$ and $\delta p(\mathbf{r}, t)$ respectively) in terms of the density and temperature fluctuations ($\delta n(\mathbf{r}, t)$ and $\delta T(\mathbf{r}, t)$ respectively) and subsequently solve this system of equations for the density fluctuations. In this way (making use of the definition in Eq. (2)) one obtains the hydrodynamic expression for the dynamical structure factor (e.g. [5],[6])

$$\begin{aligned} \frac{S(k, \omega)}{S(k)} &= \frac{\gamma - 1}{\gamma} \frac{2D_T k^2}{\omega^2 + (D_T k^2)^2} \\ &+ \frac{1}{\gamma} \left[\frac{\sigma k^2}{(\omega + c_s k)^2 + (\sigma k^2)^2} + \frac{\sigma k^2}{(\omega - c_s k)^2 + (\sigma k^2)^2} \right] \end{aligned} \quad (4)$$

In Eq. (4), γ is the ratio of heat capacities (C_v/C_p), $D_T = \kappa/(nC_p)$ is the thermal diffusivity, c_s the adiabatic sound speed, and σ the sound attenuation coefficient defined by

$$\sigma = \frac{1}{2} \left[\frac{(\gamma - 1)}{\gamma} \frac{\kappa}{nC_v} + \frac{4}{3} \frac{\eta + \zeta}{nm} \right]$$

Since the entire physics of the Yukawa model depends only on the coupling strength and range, so each of these four unknowns (γ , c_s , σ and D_T) appearing in Eq. (4) are functions of Γ and r_c^* only. Indeed, since the sound speed and heat capacities are known to reasonable accuracy from equation of state calculations [7], and the viscosities and thermal conductivity have also been calculated [8],[9], the macroscopic description of Eq. (4) is effectively known – as

required for our study – as a function of k and ω . We note that the equation of state calculations show that $\gamma \approx 1$ for the Γ and r_c^* values of interest here; therefore the hydrodynamic description of the DSF as given in Eq. (4) reduces to a single 'Brillouin' peak whose position in frequency space is given by $\omega = c_s k$ with a height and width that scale as $1/k^2$ and k^2 respectively. It is these predictions of the macroscopic description that we wish to test against the 'exact' DSF as computed with the MD simulations.

Range of Applicability of the Hydrodynamic Description

Fig. 1 shows $S(k, \omega)$ as calculated from MD plotted alongside Eq. (4) for several choices of coupling strength and range.

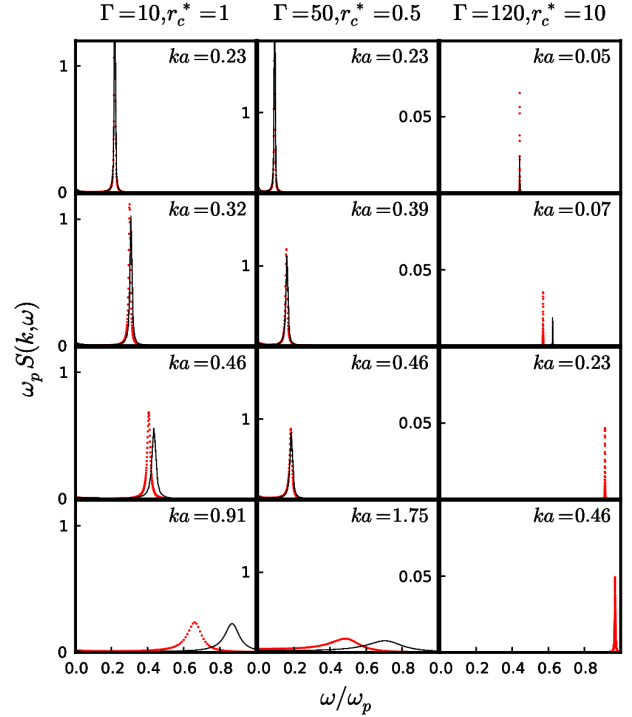


Figure 1: DSF as calculated from MD (red dots) compared with the DSF given by the hydrodynamic description in Eq. (4) (black line) for three different Γ and r_c^* values.

As shown in Fig 1. (top three panels) we find that one can always go to lengthscales sufficiently long (i.e. sufficiently small k) that the hydrodynamic description is valid. We find that rather than requiring conditions on both k and ω , the hydrodynamic description is valid providing $ka < (ka)_{max}$, where $(ka)_{max}$ is the wavenumber beyond which the hydrodynamic description breaks down (i.e. no longer works well). Below $(ka)_{max}$, the hydrodynamic description of $S(k, \omega)$ works very well indeed for all ω – the height, width and position of the Brillouin peak are all predicted accurately. Physically, no condition on ω is required because the system dynamics at sufficiently long lengthscales are always slow enough for the macroscopic equations to be applicable (this is illustrated in Fig 1. – the Brillouin peak is always at small ω for small k). As can also be seen in Fig 1., at k values greater than $(ka)_{max}$ the Brillouin peak is always located at smaller ω than predicted by the hydrodynamic description ($\omega = c_s k$) – this is a well known phenomenon in real liquids (see e.g. [10]). The manner in which the excellent correspondence between the hydrodynamic description and the true microscopic dynamics of the system breaks down is shown for the height and width of the peak in Fig. 2. As seen from Fig. 2, the true height of the DSF

decreases more slowly as k increases than Eq. (4) predicts, and the true width of the DSF also increases more slowly.

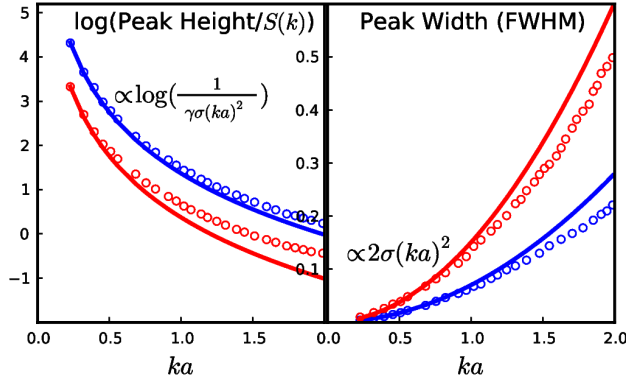


Figure 2: Height and width of Brillouin peak as computed from MD (open circles), and the predictions of the hydrodynamic description (solid lines). Red: $\Gamma=10, r_c^*=0.5$, Blue: $\Gamma=50, r_c^*=0.5$.

By increasing the importance of many body correlations in the system (i.e. increasing Γ) for a specific r_c^* value, we find that $(ka)_{max}$ is not in general significantly changed (not shown). This is perhaps a slightly surprising result, as the system intuitively becomes more 'collisional' as Γ increases, and one may therefore expect $(ka)_{max}$ to increase with increasing Γ . On the other hand, the range r_c^* is of the utmost consequence to the domain of validity of the hydrodynamic description: the hydrodynamic description is only valid on lengthscales greater than this range. This is most effectively illustrated by looking at the position of the Brillouin peak for a selection of r_c^* values (Fig. 3). As can be seen in Fig. 3, when the interaction potential becomes more long ranged, it is necessary to look at increasingly long lengthscales (small ka) for the hydrodynamic description to be valid.

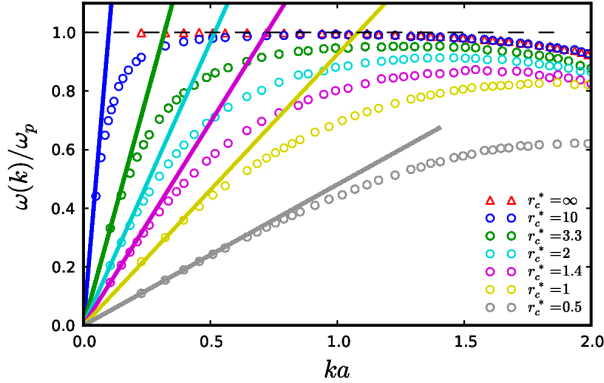


Figure 3: Dependence of the domain of validity of the hydrodynamic description on the range of the potential. The open circles are the MD results for the position of the Brillouin peak, with the associated linear relations (solid lines) $\omega=c_s k$ also shown for each r_c^* value. In all cases $\Gamma=10$.

Indeed, in terms of the range of the potential rather than the inter-particle spacing, we find from a fit to the MD results the approximate relation

$$(ka)_{max} \sim 0.39(r_c^*)^{-0.86} \quad (5)$$

Concluding Remarks

Yukawa potentials are frequently used in dense plasma physics [3]. Recently, it was shown that the use of a Yukawa potential to describe the effective ion-ion interaction, with a range given by the electron-ion screening length in the linear approximation, was able to reproduce well the results of *ab-initio* density functional theory (DFT) simulations [11]. Therefore, within the range of applicability given approximately by Eq. (5), the hydrodynamic description can provide an important complement to DFT techniques, as no dynamical information is available in those simulations.

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The Accuracy of Rear-Surface Measurements of the Angular-Divergence of a Laser-Generated Fast-Electron Beam

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Introduction

The interaction between high-intensity lasers and solid targets is of key importance in fast-ignition inertial confinement fusion [1] and ion-acceleration [2]. Cone-guided fast-ignition uses the interaction between a high-intensity heater beam and the inner tip of a solid gold cone to generate fast-electrons which then go on to heat a compressed pellet of fusion fuel. The angular divergence of the electron beam is critical in determining the flux of fast-electrons reaching the core and so their absorption there. In laser-solid target experiments relevant to this fusion scheme the angular divergence is usually inferred from temperature measurements at the rear-surface or from the size of the ion-emitting region [3-6]. In target-normal sheath ion-acceleration (TNSA), which has potential applications to cancer-therapy, a beam of ions is accelerated by a sheath electric field at the rear-surface. This field has been observed in experiments to spread very rapidly along the target rear-surface, giving a large ion-emitting region [7].

The validity of inferring the angular divergence of the fast-electron beam from rear-surface measurements will be discussed in detail; in particular the temperature measurement will be shown to give a good estimate of the beam divergence only when the re-circulation of electrons (i.e. their reflection by the sheath fields at the target edges causing them to bounce laterally down the target) was suppressed. The size of the ion-spot will be shown to be a very poor way of estimating the divergence.

Simulation results

Solid density plasmas were simulated with the hybrid Vlasov code FIDO [8]; the Vlasov equation was solved for the fast-electrons generated by the high-intensity laser and a fluid treatment used for the cold electrons which made up the target. The Vlasov equation was solved in 4D (x, y, p, θ) for the evolution of the electron distribution function in phase space. A solid aluminium target was simulated, with density $6 \times 10^{28} \text{ m}^{-3}$ and a thickness of 10 microns. The laser intensity was $2 \times 10^{19} \text{ W cm}^{-2}$ with an absorption fraction of 0.3. The laser spot was Gaussian with width 3.6 microns. The electron beam was injected moving in the +y-direction. Re-circulating currents could be suppressed by stretching the numerical grid in the -y direction, allowing the fast-electrons to escape.

Fast-electron number density and background temperature profiles are often used to infer the width of the beam. Figure 1 shows that fast-electron refluxing significantly broadens both, but that the fast-electron number density provides a better estimate of the beam size (20 microns). The temperature is broadened by the rapid saturation of the Ohmic heating rate, as illustrated by equation (1). However, when refluxing was suppressed the width of the temperature profile gave a good estimate of that of the beam.

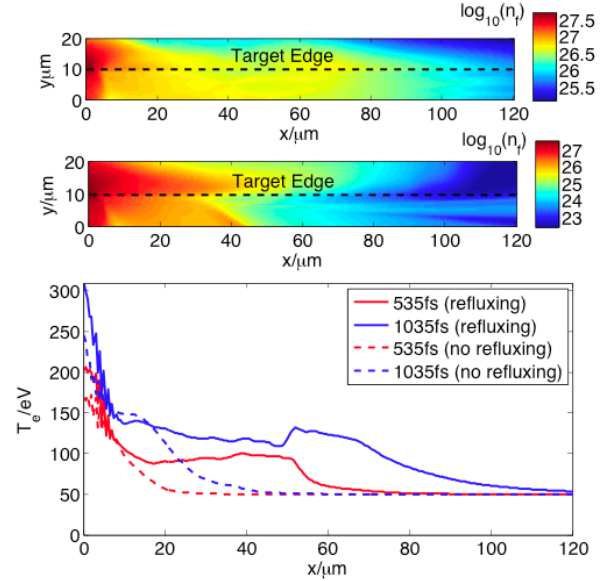


Fig.1: Fast-electron number density after 1035fs with refluxing (top) and without refluxing (middle) after 1035fs. Background temperature at the rear-surface (bottom).

$$\frac{\partial T_e}{\partial t} \propto T_e^{-3/2} \quad (1)$$

When the fast electrons reached the rear surface of the target and entered the vacuum they generated a sheath electric field in the y-direction. Experimentally, sheath fields are seen to move very rapidly along the target's rear surface, therefore there must be a fast-electron current over a large area on the rear-surface. This has so far been explained by re-circulation leading to electrons present far from the injection region.

The width of the sheath region, measured by the width of the ion-emitting region on the rear of the target, yields a very poor measure of the beam divergence due to the insensitivity of the sheath field to the number of incoming electrons. In the simplest model of a sheath the product of the peak electric field (E_y^{peak}) and the sheath width (which is set by the Debye length of the fast-electrons λ_{Df}) must be approximately equal to the average energy of the incoming fast-electrons. It is therefore straightforward to show that the sheath potential (ϕ) is independent of the number density of incoming fast electrons (n_f) and that the peak field scales as $n_f^{1/2}$, as shown in equation (2). This is borne out by the profiles at the rear of the target after 100fs, as shown in figure 2.

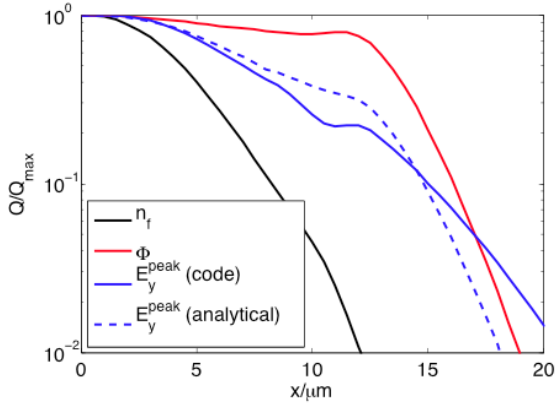


Fig.2: Fast-electron number density, peak sheath field and sheath potential along the target's rear-surface after 100fs, and $E_y^{\text{peak}} \propto n_f^{1/2}$.

$$E_y^{\text{peak}} = \frac{\phi}{\lambda_{\text{df}}} = \phi \left(\frac{e^2}{\epsilon_0 \phi} \right)^{1/2} n_f^{1/2} \quad (2)$$

The Ampere-Maxwell law gives the rate of growth of the sheath field.

$$\frac{\partial E_y}{\partial t} = -\frac{j_y}{\epsilon_0} = \frac{en_f c \cos \theta}{\epsilon_0} \quad (3)$$

This can be integrated assuming n_f is constant, until E_y grows so that it may reflect a significant number of fast-electrons. Therefore, in regions of the target where there are few fast-electrons, the sheath field and potential are proportional to n_f . This explains the fall-off of these quantities far from the beam in figure 2.

The speed of expansion can be explained by noting that the insensitivity of the sheath field to n_f meant that relatively low current densities (<1% of the maximum) generated large fields. Initially, the field was generated by electrons on their first pass through the target. In this case the spreading of the sheath field along the rear-surface was due to the difference in arrival times for electrons with different injection angles; those injected at large angles to the target normal had further to go before hitting the rear-surface; this is illustrated in figure 3. In this case the speed of lateral expansion of the sheath field (v_{exp}), assuming the electrons were travelling at the speed of light and that they generated a sheath as soon as they arrived at the rear-surface, is given by equation (4).

$$v_{\text{exp}} = \frac{c}{\sin \theta} \quad (4)$$

Where θ is the angle from the target normal to the position on the rear of the target. Therefore the field expanded super-luminally. Note that this does not violate special relativity; information was not being transferred from the centre of the rear surface to the point at which the sheath had reached but from the injection region to this point at c .

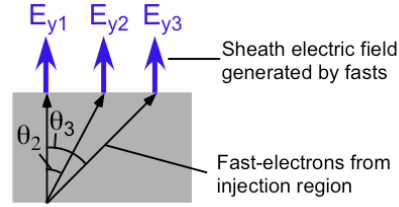


Fig.3: An illustration of the cause of sheath field spreading. At time τ_1 electrons with an incident angle of 0° generate E_{y1} , some time later (τ_2) those with angle θ_2 generate E_{y2} . Finally those with θ_3 generate E_{y3} .

As a corollary, this weak dependence means that it may be possible to increase the efficiency of TNSA ion acceleration by defocusing the beam somewhat in order to scatter the electrons as widely as possible in the target (provided their energy is not curtailed too much by the resultant drop in laser intensity).

Conclusions

Experimental measurements of the angular divergence of fast-electron beams generated in short-pulse laser-solid target interactions may be misleading unless analysed carefully. Although the fast-electron number density was strongly peaked in the region of the beam, re-circulating currents significantly widened the temperature hot-spot on the back. The size of the sheath potential on the back was weakly dependent on the size of the electron beam inside the target and so a measurement of the size of the ion-emitting region gave a very poor estimate of the angular divergence. The sheath field's expansion was controlled by electrons on their first pass through the target and so could expand super-luminally.

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High Absorption of Ultra-Intense Laser Pulses in Near Critical Plasmas

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Introduction

Of all the problems that ultra-intense laser-plasma physics is concerned with, the coupling of energy from the laser pulse into the kinetic energy of the plasma particles is perhaps the most important. Attaining efficient coupling is highly important and even critical to a number of prospective applications. Fast Ignition ICF and laser-driven ion acceleration are examples of such applications where coupling efficiency is either a very important or critical issue.

In the work reported here, we analyzed the absorption of 30-100fs, $\lambda \sim 1\mu\text{m}$, $a_0 \gg 1$ laser pulses in near-critical plasmas. By this we mean plasmas are relativistically transparent and span the density range $0.1n_c < n_e < a_0 n_c$, where n_c is the non-relativistic critical density. The objective of this study is to understand the *gross* properties of these interactions – the propagation velocity of the pulse, the rate of energy absorption, and the scaling of the fast electron energy. This parameter range is particularly interesting as very high absorption (up to 70%) is possible.

Previous Work

The general problem of absorption in underdense plasmas has been the subject of a number of previous studies. Much of this has mainly been concerned with plasmas that are below $0.1n_c$. In this work we have been particularly concerned with Leading Edge Depletion which was originally described in a number of papers (e.g. [1,2]), but here we make particular reference to the paper by Decker and co-workers [1]. The absorption of laser energy via transverse expulsion of electrons is a concept that is described in a number of papers, see for examples.

The purpose of this work was to re-examine these two concepts – Leading Edge Depletion (LED) and Transverse Ponderomotive Acceleration (TPA) – and to obtain basic analytic models for their gross properties that are good descriptions of what is observed in numerical simulations. Part of this is to determine the answer to an even more fundamental question – are these regimes genuinely distinct at all?

Leading Edge Depletion Theory

Decker et al. give the leading edge propagation velocity as,

$$\frac{u}{c} = 1 - \frac{n_e}{n_c}, \quad (1)$$

in the limit of $n_r \ll n_c$. The problem with this expression is that in the region $n_c < n_r < a_0 n_c$ one obtains a negative velocity which is at odds with numerical simulations. We set out to derive a better analytic expression by using energy conservation arguments.

In LED absorption notionally occurs at the leading edge by electrons overtaking the leading edge (which travels at a velocity less than c) and taking a momentum of $a_0^2 m_e c/2$ with them. This means that the laser pulse must lose energy at a rate of,

$$\frac{dE}{dt} = -\frac{a_0^2}{2} m_e c^2 n_e u, \quad (2)$$

And the loss of energy manifests as an erosion of the laser field that propagates backwards at velocity v_{er} . These velocities are related via $u = v_g - v_{er}$, where v_g is the group velocity of the laser pulse. This implies that,

$$\frac{dE}{dt} = -\frac{v_{er} I}{c}, \quad (3)$$

where the laser intensity relates to a_0 via $I = a_0^2 m_e c^2 n_c$. Equating 2 and 3 yields the following expression,

$$u = \frac{v_g}{\frac{n_e}{n_c} + 1}. \quad (4)$$

In the limit of $n_e \ll n_c$ one retrieves Decker's expression by taking the limit of eqn. 4. In LED the fast electron energy should scale as a_0^2 .

Transverse Ponderomotive Acceleration Theory

In 2D and 3D the other route for absorbing energy is transverse ponderomotive expulsion. If we note that each electron expelled will take an energy of $a_0 m_e c^2$ from the laser pulse then we can repeat the LED analysis except with,

$$\frac{dE}{dt} = -a_0 m_e c^2 n_e u, \quad (5)$$

, and obtain the following expression,

$$u = \frac{v_g}{\frac{n_e}{a_0 n_c} + 1}. \quad (6)$$

In TPA, the fast electron energy should scale as a_0 .

1D PIC simulations

1D electromagnetic PIC simulations were carried out to validate the theory of LED absorption in 1D. This was done with a $50\mu\text{m}$ plasma and with a 40fs 'flat-topped' laser pulse that was circularly polarized and had a wavelength of $1\mu\text{m}$. In one set of simulations we set $a_0 = 10$ and varied the plasma density over the range $0.5-2n_c$ and measured the leading edge propagation velocity. In another set, n_e was set to $0.6n_c$, a_0 was varied, and the scaling of the fast electron energy was measured. The results are summarized in figure 1 where we show that eqn. 4 and the a_0^2 scaling of LED are in excellent agreement with the simulation results.

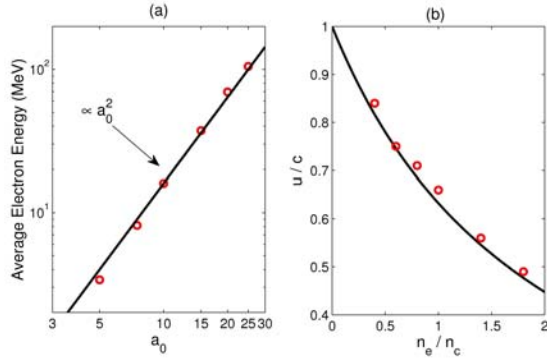


Figure 1. (a) Test of energy scaling in 1D PIC simulation, (b) Test of leading edge propagation velocity in 1D PIC simulation. Black lines correspond to LED analytic results (eqn. 4). Red circles are simulation results.

2D PIC simulations

Next we used 2D PIC simulations using the EPOCH code to study 2D situations where both LED and TPA might occur. In these simulations we also used a $50\mu\text{m}$ thick plasma and a ‘flat-topped’ circularly polarized laser pulse. The laser pulse had a Gaussian transverse distribution, characterized by r_L .

We can summarize the results of this investigation by looking at the plots shown in figures 2 and 3.

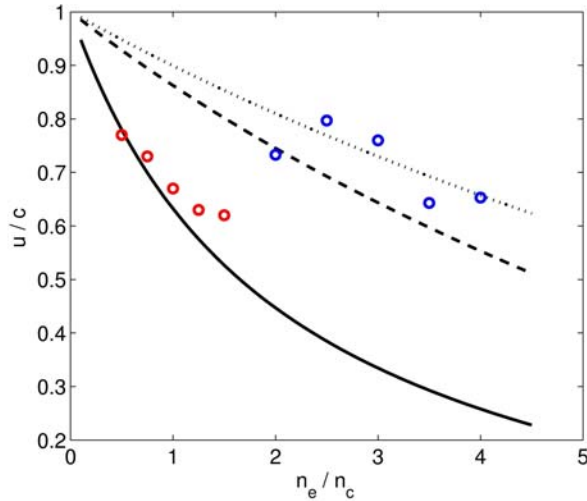


Figure 2. Plot of leading edge velocity, u , from 2D PIC simulations (red circles and blue circles). Both sets have a pulse with $a_0 = 10$ and $r_L = 10\mu\text{m}$. The solid black line corresponds to eqn. 4 (LED) and the dashed line corresponds to eqn. 6 (TPA). The dotted line corresponds to eqn. 6 (TPA) but with $a_0^2/2$.

Figure 2 shows the leading edge velocity that was observed over a range of simulations on varying the plasma density. In these two sets, $a_0 = 10$ and $r_L = 10\mu\text{m}$. Figure 2 shows that the leading edge velocities closely follow either one of the two curves, indicating that there is a LED regime that is well described by the analytic model and a TPA regime that is also well described by the analytic model. The intensity correction included by the dotted line in figure 2 is put there because of intensity increases due to the strong filamentation of the laser pulse that occurs in the TPA regime. Figure 2 also shows that the two regimes are *distinct*, i.e. one does not really observe an admixture of the two, except for regions where there is a smooth transition between the two regimes.

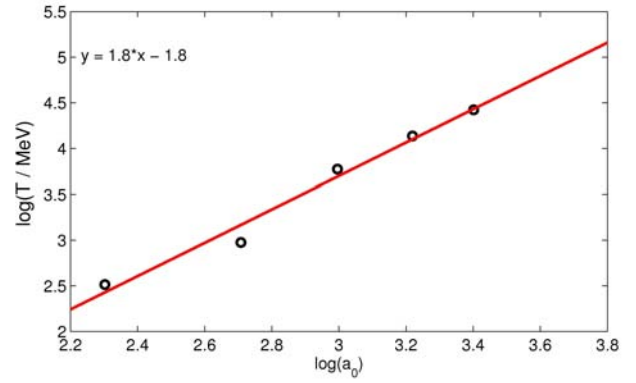


Figure 3. Log-log plot of the fast electron temperature against a_0 for a set of simulations with $n_e = 0.5n_c$ and $r_L = 10\mu\text{m}$. This shows a power law scaling of $a_0^{1.8}$ which is very close to the LED scaling of a_0^2 .

Figure 3 shows the results of one check of the fast electron energy scaling with a_0 . At low density where we expected the LED regime to apply we found a scaling of $a_0^{1.8}$ which is very close to the LED scaling of a_0^2 . At high densities we observed a scaling of $a_0^{1.1}$ which is very close to the TPA scaling of a_0 . At an intermediate density we observed a scaling of $a_0^{1.3-1.4}$, which lies between these two cases.

Overall we found that the 2D PIC simulations showed the existence of separate LED and TPA regimes with smooth transitions between them. In these distinct regimes, the gross properties are well predicted by the new analytic formulae that we have derived.

Conclusions

Two general ideas about how absorption occurs in near-critical plasma have existed for a number of years. In this body of work we have critically examined both ideas and derived new analytic formulae to describe the gross properties that improve on the existing formulae. Our simulations show that both ideas, Leading Edge Depletion and Transverse Ponderomotive Acceleration, can exist as distinct regimes and in these regimes the analytic formulae provide good predictions in comparison to the simulation results.

Acknowledgements

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Modelling Spitzer transport in laser produced plasmas by direct Vlasov methods with a BGK collision operator

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Introduction

Collisional processes are of critical importance to short-pulse laser matter interaction. The absorption of incident laser energy is dominated by collisionless mechanisms^{1,2,3}. However, the onward transport of the flux of fast moving, essentially collisionless, electrons and the resultant heating of the target is dependent on the response of the cold, resistive, background plasma.

Considerable progress has been made in modelling transport phenomena in both long and short pulse laser plasma interactions using an approach to the Vlasov-Fokker-Planck problem based on a decomposition of the electron distribution function into spherical harmonics, or a similar set of basis functions^{4,5}. However, for modelling the laser-plasma interaction region directly a direct Vlasov solver is desirable: unfortunately, solving the VFP problem on the discrete phase space grid of a direct Vlasov solver is extremely demanding, computationally speaking⁶.

Here we briefly outline a simple approach to including collisional physics in the direct Vlasov solver VALIS⁷, based on the BGK (Bhatnagar, Gross and Krook) collision operator, which does not have the computational overheads associated with the VFP approach. We are able to demonstrate the efficacy of this approach in two key transport problems in laser plasma interaction: thermal conduction and electrical resistivity.

The Bhatnagar, Gross and Krook collision operator

Collisional effects form the right hand side of the Boltzmann equation but are absent from the Vlasov equation. They can be included in a Vlasov solver, such as VALIS, by the addition of an appropriate collision operator, so that for species j :

$$\frac{\partial f_j}{\partial t} + \frac{\mathbf{u}}{\gamma} \cdot \nabla_{\mathbf{x}} f_j + \frac{q_j}{m_j} \left(\mathbf{E} + \frac{\mathbf{u}}{\gamma} \times \mathbf{B} \right) \cdot \nabla_{\mathbf{u}} f_j = C_j(f_j) \quad (1)$$

where C represents the collision operator:

$$C_j(f_j) = \sum_k \left(\frac{\partial f_j}{\partial t} \right)_{C,k} \quad (2)$$

and $\left(\frac{\partial f_j}{\partial t} \right)_{C,k}$ represents the change in f_k due to collisions

with species k . Any collision operator should have the following characteristics: it must conserve mass, momentum and energy; it must have an H -theorem (that is to say it must asymptotically approach a unique equilibrium distribution in a homogenous plasma, the Maxwellian distribution); and it must maintain positive f . Here we consider and implementation of a simple BGK collision operator⁸:

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$$\left(\frac{\partial f_j}{\partial t} \right)_{C,k} = -\nu_{jk} (f_j - F_j) \quad (3)$$

where ν_{jk} is the collision frequency for collisions between species j and k , F_j is the equilibrium distribution: Maxwell-Boltzmann in the non-relativistic limit and Maxwell-Jüttner in the relativistic case.

Following the treatment devised by Mannheimer et al.⁹ we use the high and low energy collision frequencies from the NRL Plasma Formulary¹⁰ analytically matched so that, for example, the electron-ion collision frequency becomes:

$$\nu_{ei} = \frac{6.43 \times 10^{-8} \ln \Lambda Z n_e}{\left(\frac{m_e}{m_i} T_i \right)^{3/2} + \frac{5.8}{7.7} E_{ne}^{3/2}} \quad (4)$$

where subscripts e and i denote electron and ion quantities respectively and E_{ne} represents the electron kinetic energy. The value of the Coulomb log is also taken from the NRL Plasma Formulary.

The BGK operator is treated implicitly so that, for electron-ion collisions only:

$$f_e^{n+1} - f_e^n = \nu_{ei} \Delta t (f_e^{n+1} - F_e) \quad (5)$$

giving:

$$f_e^{n+1} - f_e^n = \frac{\nu_{ei} \Delta t (f_e^n - F_e)}{(1 + \nu_{ei} \Delta t)} \quad (6)$$

By taking velocity moments of Equation 6, it is clear that the BGK operator can be made to conserve energy, mass and momentum by a suitable choice of F . For the electron-ion collision term to have the correct conservation properties at each time-step we find the F_e which satisfies:

$$\int \left\{ \frac{\nu_{ei}}{1 + \Delta t \nu_{ei}} \left(\frac{u_e}{\gamma} \right)^m (f_e^n - F_e) \right\} du = 0 \quad (7)$$

where $m = 0, 1$ or 2 . Since the form of F_e is known we can simply calculate the density, drift speed and temperature of F_e iteratively. For the tests detailed here, we consider only the non-relativistic limit, fixing $\gamma = 1$, and do not impose conservation of momentum for any electron-ion collisions.

This implementation of the BGK approach has a number of desirable properties: it can be implemented in a fully implicit and conservative manner and implemented independently of the solution of Vlasov's equation; it can be applied to multi-species systems; the impact of collisions or changes to the underlying

physics can be easily assessed through changes to the calculation of the collision frequencies; and it does not rely on an expansion in the reciprocal of the Coulomb log.

This is potentially a practical way of including collisional effects in a direct Vlasov solver like VALIS. Inclusion of the full Landau form of the Fokker-Planck equation in an Eulerian Vlasov solver can be achieved⁶, but at significant computational cost, which can make the application of such approaches impractical with the level of HPC resources currently available.

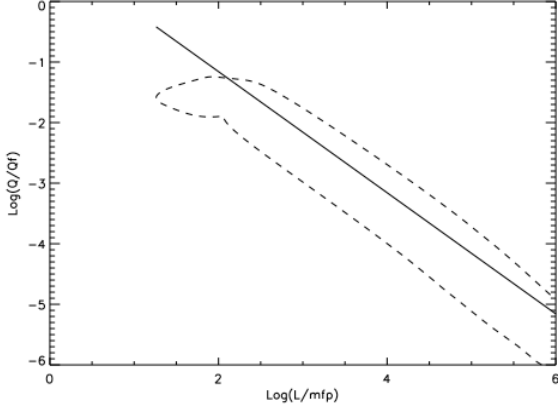


Figure 1. Ratio of calculated electron heat flux (Q) to the free streaming limit (Q_f) as a function of scale-length over electron mean free path. The solid line is the Braginskii (Spitzer-Harm) predicted heat flux. The dashed line is the computed heat flux.

Thermal conductivity test

We initialise the system with a tanh profile for the electron temperature (from 400 eV down to 100 eV), stationary ions ($Z=4$) and an electron number density of $2.97 \times 10^{26} \text{ m}^{-3}$ so that there are 100 particles per Debye sphere for the 100 eV electrons. The computed heat flux compared to Spitzer-Harm is plotted in Figure 1 after 1000 hot electron plasma periods. This shows heat flux limiting of ~ 0.1 . The computed heat flux is multi-valued when plotted in this way, the lower section of the dashed curve corresponds to the hot side of the temperature profile and exhibits thermal flux limiting. The upper section corresponds to the cold side of the simulation and exceeds the Spitzer-Harm value, given by a solid line, due to the contribution from hot electrons from the higher temperature region. These results are in qualitative and quantitative agreement with previous simulations based on the full Fokker-Planck collision operator¹¹.

Plasma resistivity

Short-pulse laser plasma interaction at the target surface drives high energy ‘hot’ electrons into the body of the target. These electrons can generally be treated as collisionless, but they draw a return current of slow-moving collisional electrons¹², which are responsible in large part for heating the target as well as influencing the development of magnetic fields, which may in turn collimate the hot electron beam¹³. Therefore, it is important that any transport model intended for application to problems in short-pulse laser produced plasmas be able to reproduce well understood phenomena such as Spitzer resistivity.

We adopt a 1D1P system (one dimension in space and one in momentum) of mobile electrons and immobile protons 2 nm in length and periodic in x , with initial electron and ion temperatures of 100 eV and with a momentum extent equivalent to $\pm 20v_{te}$, where v_{te} is the electron thermal velocity. A relatively coarse phase space resolution of 8 cells in x by 64 cells in u is utilised throughout. We choose an electron number density of 10^{29} m^{-3} and apply a fixed external electric field of between 10^7 and 10^9 Vm^{-1} , while disabling the code’s Maxwell field solver.

The average current in x versus time can then be compared with the expected value:

$$J_{SH} = \frac{E}{\eta_{SH}} \quad (8)$$

where, considering only electron-ion collisions^{14, 15}:

$$\eta_{SH} = \frac{\alpha_e m_e}{e^2 n_i \tau_{ei}} \quad (9)$$

and the collision time, is given as:

$$\tau_{ei} = 3\pi^{3/2} \epsilon_0^2 \sqrt{m_e} \frac{(2k_B T_e)^{3/2}}{e^4 Z^2 n_i \ln \Lambda} \quad (10)$$

Here α_e is the classical transport coefficient calculated in Ref. 15 from the numerical solution of the Fokker-Planck equation and all quantities are given in SI units.

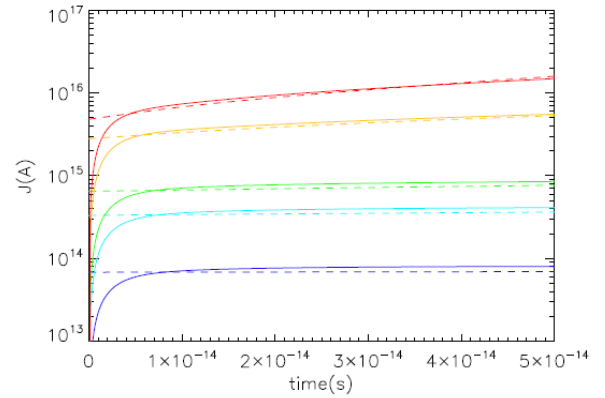


Figure 2. Comparison of Spitzer currents (dashed lines) with those calculated using VALIS with a BGK collision operator (solid lines) for external fields of: 10^7 Vm^{-1} (blue); $5 \times 10^7 \text{ Vm}^{-1}$ (cyan); 10^8 Vm^{-1} (green); $5 \times 10^8 \text{ Vm}^{-1}$ (yellow); and 10^9 Vm^{-1} (red), for the case of electron-ion collisions only. The gradient of the lines is a result of Ohmic heating.

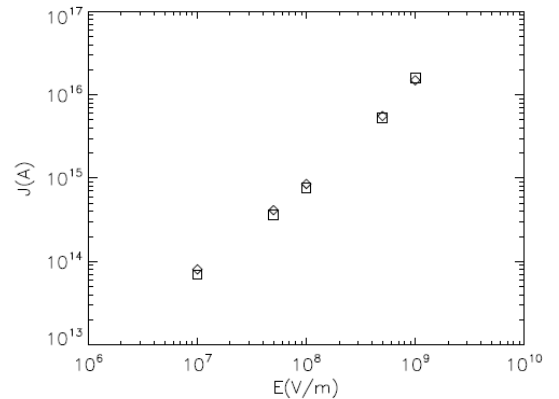


Figure 3. Comparison of Spitzer currents (squares) with those calculated using VALIS with a BGK collision operator (diamonds) after 50 fs for a range of applied fields and including only electron-ion collisions.

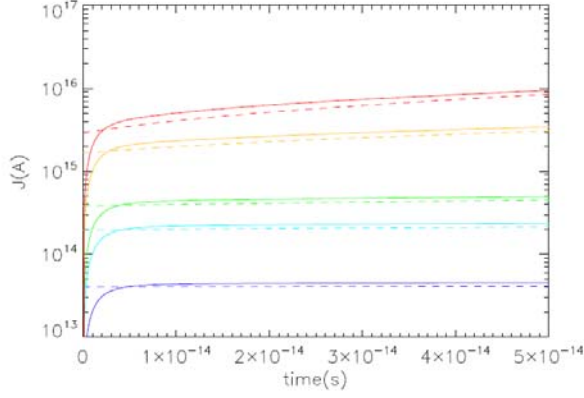


Figure 4. Comparison of Spitzer currents (dashed lines) with those calculated using VALIS with a BGK collision operator (solid lines) for external fields of: 10^7Vm^{-1} (blue); $5 \times 10^7 \text{Vm}^{-1}$ (cyan); 10^8Vm^{-1} (green); $5 \times 10^8 \text{Vm}^{-1}$ (yellow); and 10^9Vm^{-1} (red), for the case of both electron-electron and electron-ion collisions.

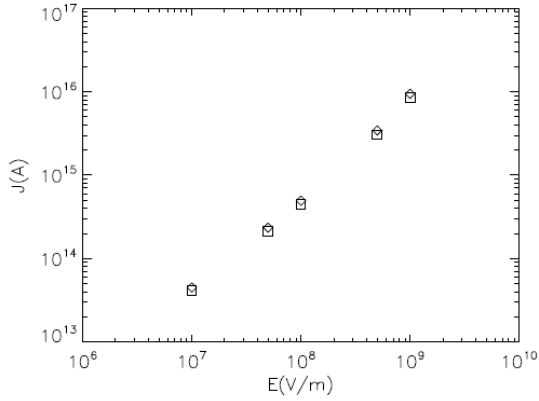


Figure 5. Comparison of Spitzer currents (squares) with those calculated using VALIS with a BGK collision operator (diamonds) after 50fs for a range of applied fields and including both electron-electron and electron-ion collisions.

Figures 2 and 3 summarise results for a range of external fields. VALIS, with the BGK collision operator is able to reproduce the expected Spitzer current across the range of fields. When the effect of electron-electron collisions (for which one must add an additional pre-factor of $\sim 16/3\pi$ to equation 9) are included, the results are equally encouraging – as shown in Figures 4 and 5. Broadly similar results have been achieved at number densities of 10^{28} and 10^{29}m^{-3} over the range of fields. As one would expect, results in the low density, high field case display a higher drift velocity and demonstrate considerably more Ohmic heating, compared to the low field, high density case, where the deviation from the initial Maxwellian is much less pronounced, see Figure 6.

Conclusions

We have outlined a simple BGK collision operator for the Eulerian Vlasov solver VALIS. This implementation can be applied to multi-species systems including both inter and intra-species collisions and is formulated so that particle number, energy and momentum are conserved. Furthermore, the fully implicit implementation allows the method to be applied independently of the scheme used to solve Vlasov's equation^{7,16}.

Two simple 1D tests demonstrate the potential of this approach for modelling problems in laser-plasma interaction, making this

approach a practical way of including collisional effects in a direct Vlasov solver like VALIS and allowing the self-consistent modelling of absorption and transport without the need to solve for the full Landau form of the Fokker-Planck equation. Ongoing work will extend this approach to higher dimensions, relativistic problems and realistic systems.

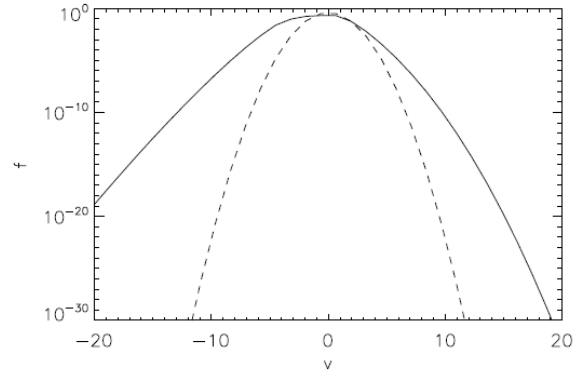
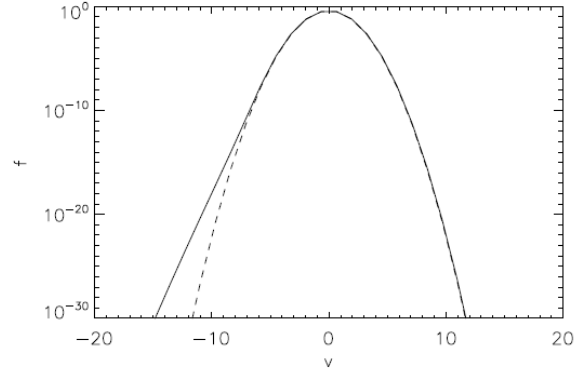


Figure 6. Initial (dashed line) and final, i.e. after 50fs, (solid line) velocity distribution functions for: an electron number density of 10^{30}m^{-3} and applied field of 10^7Vm^{-1} (top); an electron number density of 10^{30}m^{-3} and applied field of 10^7Vm^{-1} (bottom). The effect of Ohmic heating is clear in the low-density, high-field case.

Acknowledgements

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Effect of channel profile evolution on laser-driven electron acceleration in plasma channels

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Introduction

Plasma-based electron acceleration [1] holds the promise to deliver electrons at multi-GeV energies while requiring only a short acceleration distance. This was demonstrated most vividly when the energy of a 42 GeV electron beam was doubled in a 0.85 m long plasma in a beam-driven wakefield experiment [2]. While operating at lower energies, laser-driven wakefield acceleration has the advantage of producing electron bunches with an energy spread below 5% [3–5]. To increase the output energy of a plasma accelerator, one has to decrease the density of the background plasma and increase the acceleration length. In the case of laser-driven acceleration, this introduces the complication of laser pulse divergence. Without special measures, the length over which the laser pulse will stay spatially confined and capable of driving a decent wakefield is much shorter than the required acceleration length. An often used technique to overcome laser pulse divergence and increase the laser-plasma interaction length is pulse guiding by means of a preformed plasma channel. In such a channel, the plasma density is lowest along the channel's central axis, so the index of refraction is highest there and the laser pulse is guided as it would be in a glass fiber. In [4], a plasma channel was created by line-focusing a secondary laser pulse onto a gas jet; this channel was then used to guide the primary laser pulse. However, for longer acceleration lengths and lower plasma densities, plasma channels formed by hydrogen-filled capillary discharge waveguides [6, 7] are much more suitable. Using a 40 TW laser pulse guided by such a channel, electron bunches have been accelerated to up to 1 GeV with 5% energy spread [8].

Review of previous experimental results

Recently, laser wakefield acceleration in a hydrogen-filled capillary discharge waveguide was studied experimentally on the Astra Ti:sapphire laser at the Rutherford Appleton Laboratory [9]. During this experiment, plasma electron trapping and acceleration was observed. The accelerated electron bunches had a mean energy of up to 200 MeV and around 15 MeV spread. In addition, it was found that the probability of electron trapping was strongly dependent on the delay between the onset of the discharge current and the arrival of the laser pulse. Electron acceleration was only observed during a narrow delay interval, when the discharge current was already starting to switch off and laser energy transmission was past its peak. Interferometric and Raman spectroscopic measurements confirmed that electron trapping coincided with a decrease of both the degree to which the hydrogen was ionised and the depth of the plasma channel, i.e. with a general decrease in channel quality. No electrons were observed when the discharge current, laser energy transmission, channel depth and degree of hydrogen ionisation were at their peak, i.e. when the channel quality was optimal.

Simulation results

From these results, it is clear that electron injection in a plasma channel depends sensitively on the state of the plasma channel. In [9], it was suggested that the degree of ionisation may have a significant effect on electron trapping: electrons that are “born” during the passage of the laser will have a different velocity than free electrons that were already there, and may therefore be more eligible for trapping by the wakefield. However, other plasma parameters evolve simultaneously with the degree of ionisation, e.g. the overall plasma density and the transverse profile of the plasma channel. While it is not possible to isolate the effect of a change in each parameter in the experiments, it is certainly possible to do so in computer simulations. To investigate the influence of the degree of ionisation, as well as the background plasma density, channel profile and laser pulse intensity, on electron injection, we have performed a series of two-dimensional (2-D) numerical simulations using the particle-in-cell (PIC) code OSIRIS [10]. This code implements the full relativistic PIC algorithms, and also includes routines to model ionisation of neutral gases (H, He, Li, Ar). A series of simulations has been performed, in which the degree of ionisation (fully ionised, 1/3 neutral gas, 1/2 neutral gas), laser intensity ($0.8 < a_0 < 2.0$), plasma background density ($n_0 = (1 - 1.5) \times 10^{19} \text{ cm}^{-3}$) and channel profile ($n(r)/n_0 = 1 + a(r/r_0)^2$, where $0 < a < 0.033$) were varied. The results of these simulations can be summarised as follows:

- Increasing the laser intensity leads to a larger wakefield amplitude, causing the injection of more electrons which are subsequently accelerated to higher energies. These results are displayed in Figure 1.
- Increasing the plasma density by only 50% leads to a significant increase in both the number of trapped electrons and their energy after a fixed acceleration distance. If the acceleration could be continued for the entire dephasing length the final energy would be higher for lower densities, but in the experimental configuration used in [9] this is not realistic because of laser pulse depletion and finite channel length. These results are displayed in Figures 1 and 2.
- Decreasing the channel depth from 0.33 to 0.165 leads to only a moderate widening of the laser spot size without loss of guiding even for intensities as low as $a_0 = 0.8$. Removing the channel altogether leads to pulse divergence, lower wakefield amplitude and a decline in particle injection for the lowest intensities ($a_0 = 0.8$). For $a_0 > 1.0$, the laser pulse is sufficiently intense to guide itself via the process of relativistic self-focusing, and removing the channel has a much smaller impact. These results are displayed in Figures 2 and 3.
- Changing the fraction of neutral gas from 0 to 1/3 or 1/2 has a small effect on the transverse profile of the laser

pulse, but no significant effect on either the shape of the wakefield or the electron injection and acceleration processes. These results are displayed in Figure 4.

Discussion and conclusions

Based on the above simulation results, the experimental results of [9] can be explained by the following mechanism. When the discharge current is at its peak and the plasma channel is fully formed, the on-axis plasma density of $(0.5 - 1.0) \times 10^{19} \text{ cm}^{-3}$ is slightly too low to trigger electron injection for the laser pulse intensity used ($a_0 \sim 0.6$). At later times, when the discharge current decreases, the plasma channel starts to “cave in”, i.e. the on-axis density increases while the channel depth decreases. This can actually be concluded from Fig. 2 of [9], which shows an increase in the total density of available electrons (free and from re-ionisation), as well as a gradual flattening of the transverse density profile. Initially, the rising on-axis density leads to a sharp enhancement of electron injection, as well as the final electron energy. Following that, the flattening density profile leads to a loss of laser pulse confinement, meaning that the laser pulse is no longer able to efficiently generate a wakefield and electron injection stops altogether. We thus find that efficient electron injection and acceleration only happens for a rather special combination of circumstances, which explains the narrow window in which accelerated electrons were observed in the experiment.

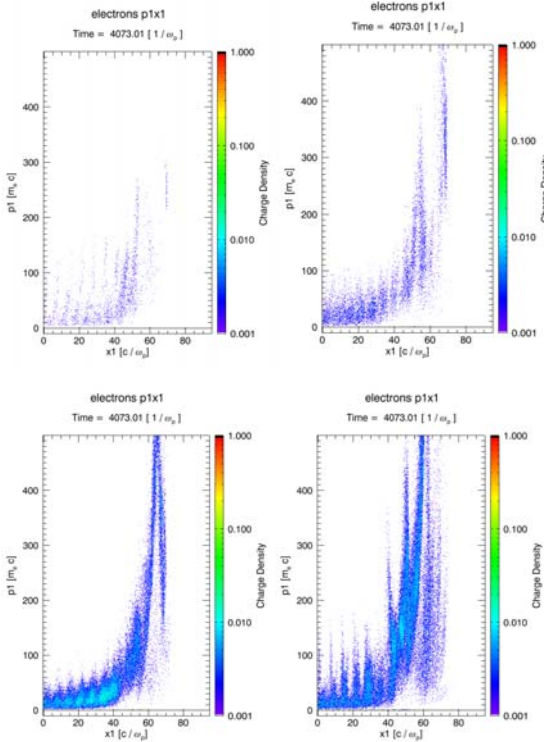


Figure 1. Electron trapping in a laser-driven wakefield, versus laser intensity. Laser amplitude a_0 is 1.19 (top left), 1.68 (top right), 2.0 (bottom left), 2.5 (bottom right), all other parameters equal. Laser spot diameter is 10 micron, wave length is 800 nm, $\omega_0/\omega_p = 13.2$, pulse duration is 50 fs. An increased laser intensity leads to enhanced electron trapping and acceleration.

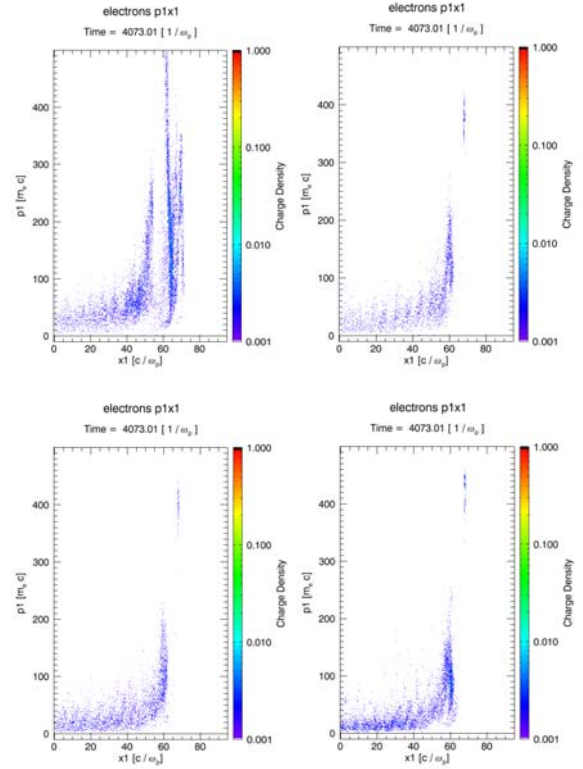


Figure 2. Top left: electron trapping for the same parameters as the top right frame of figure 1, except that $\omega_0/\omega_p = 10.56$, i.e. higher plasma density, leading to enhanced trapping and acceleration. Other frames: electron trapping versus channel depth for $a_0 = 1.19$ and $\omega_0/\omega_p = 10.56$. Channel depth is 0.33 (top right), 0.165 (bottom left) or 0 (bottom right). Reducing the channel depth leads to a decrease in electron energy, but self-focusing ensures that some electrons are trapped and accelerated even in the absence of a channel.

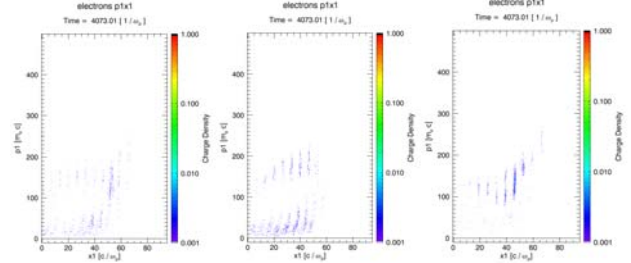


Figure 3. Electron trapping versus channel depth, for $a_0 = 0.8$. Removal of the channel inhibits wave breaking of the wakefield, leading to trapping of electron bunches at every wakefield period rather than a single bunch directly behind the laser pulse. This also has a detrimental effect on the energy spread.

In a separate simulation using a fully ionised plasma with a rather high temperature of 10 keV [11], it was found that the propagation mode of the laser pulse was different from propagation in a cold plasma, with the pulse in the cold plasma having two side lobes containing a significant fraction of the energy, while the pulse in the hot plasma had all its energy confined to one central lobe. Also, enhanced electron trapping was observed for the hot plasma. It was then suggested that re-ionisation of a partially recombined plasma by a passing laser pulse would lead to a fraction of electrons having a much higher temperature than the previously present free electrons, which would once again lead to better laser pulse confinement and enhanced electron trapping. However, while the simulations using a mixture of plasma and neutral gas did show a slight change to the transverse pulse profile on several occasions, this was not expressed in either a significantly higher electron

temperature or enhanced electron trapping. We therefore conclude that the enhanced electron trapping observed in the hot plasma entirely results from the artificially high plasma temperature. Indeed, it is well-known that wave breaking in warm plasma happens at a lower field amplitude than in cold plasma [12], while a small fraction of “thermal” electrons in a cold bulk plasma will still considerably lower the field threshold for electron trapping even if the wave itself does not break [13]. Combining Akhiezer and Polovin’s equations for cold, relativistic plasma waves [14] and Ruth and Chao’s Hamiltonian approach to quasi-static electron dynamics [13], we find the following expression for the wakefield amplitude necessary to trap an electron with initial forward speed v_i :

$$E_t \approx E_{wb} \left[1 - (\gamma_\phi v_i/2)^{1/2} + (\gamma_\phi v_i/4) \right], E_{wb} = \sqrt{2(\gamma_\phi - 1)}$$

Here, E_{wb} denotes the cold, relativistic wave-breaking field [14]. For $\gamma_\phi = 18$ and “thermal” electron energies of 1, 10, 100 and 1000 eV, we find that E_t/E_{wb} equals 0.87, 0.79, 0.66 and 0.53, respectively. This confirms that one can enhance electron trapping by increasing the plasma density and/or temperature, while changing the degree of ionisation does not really make an impact because the effect of that parameter on either density or temperature is simply too limited.

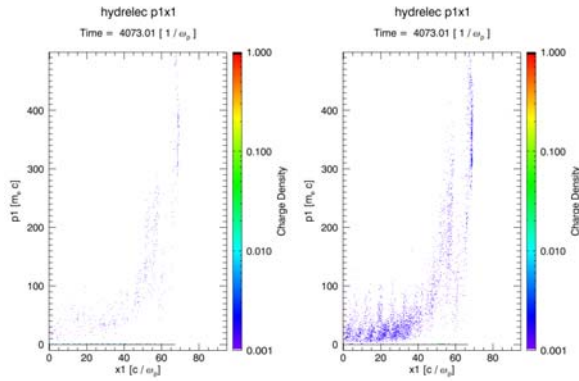


Figure 4. Energy spectrum of plasma electrons produced by ionisation processes in a partially ionized plasma: neutral fraction is 1/3 (left) or 1/2 (right). Other parameters are similar to the simulation depicted in the top right corner of Figure 1. Naturally the absolute number of electrons produced by ionisation increases with the fraction of neutrals, but their relative energy spectrum does not change much with the fraction of neutrals. The fraction of neutrals did not appear to produce qualitative changes in the energy spectrum of pre-existing electrons either.

Acknowledgements

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A Vlasov-Fokker-Planck Code For Shock Ignition

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A 2D3P parallel object-oriented Vlasov-Fokker-Planck code that relies on the expansion of the electron distribution function to spherical harmonics has been developed, in order to study non-local electron transport for Shock Ignition. The code makes use of a rigorous formalism for the collisions between electrons, which derives from the Rosenbluth potentials and conserves energy and number. This code makes it possible to accurately model the kinetic as well as the hydrodynamic behaviour of the plasma and is particularly efficient for collisional plasmas. For Shock Ignition the electron temperatures range from more than 100keV to 10eV while densities range from less than critical to greater than solid. Shock Ignition is therefore an excellent candidate for this VFP code, because the target is sufficiently collisional to allow for extremely efficient modelling.

Introduction

The ability of hot electrons to transfer energy over large distances arises from the most fundamental properties of ionized matter and leads to complex and unpredictable phenomena. Understanding and controlling non-local electron transport is crucial for Shock Ignition [1-2], which aims to increase the energy gain for Inertial Confinement Fusion (ICF) [3] without major modifications to conventional ICF designs.

In Shock Ignition, hot electrons generated near the critical surface, due to a spike in the drive laser pulse, need to distribute the energy around the compressed pellet so as to launch a spherically convergent shock. The transport of this large population of hot electrons through and around the dense plasma is associated with the generation of strong electromagnetic fields, which in turn are determined by the plasma conductivity. In this regime the Spitzer conductivity does not apply and the correct kinetic description of the total electron distribution must be considered.

One can describe the evolution of the electron distribution function with the Vlasov-Fokker-Planck equation thereby retaining the kinetic treatment of the entire electron population:

$$\frac{\partial}{\partial t} f + \mathbf{u} \cdot \frac{\partial}{\partial \mathbf{r}} f + q(\mathbf{E} + \frac{\mathbf{u}}{c} \times \mathbf{B}) \cdot \frac{\partial}{\partial \mathbf{p}} f = \left(\frac{\partial f}{\partial t} \right)_{ei} + \left(\frac{\partial f}{\partial t} \right)_{ee}$$

where $f(\mathbf{p}, \mathbf{r}, t)$ is the electron distribution function and the terms $(\partial f / \partial t)_{ei}$ and $(\partial f / \partial t)_{ee}$ describe the contributions of electron-ion and electron-electron collisions respectively. The Fokker-Planck (FP) equation resembles a Taylor expansion for small-angle deflections in velocity space and assumes linear addition of the effects of all particles within a Debye length (λ_D). Because of this cutoff and the neglect of high-order terms the FP description implies a large number of particles (N_D) within a Debye sphere and fails for very high densities and very low temperatures. There is, nevertheless, a large volume of plasma between the laser-plasma interface and the ultra-high density region, which is described well by the FP equation. In the plasma corona there are more than 100,000 electrons per Debye sphere, while in the high-density plasma their number drops to 10-100, e.g. for $T=300\text{eV}$ and $n=10^{23}/\text{cc}$ $N_D \sim 30$.

The main difficulty in solving the Vlasov equation numerically comes about from the 6D nature of the electron distribution

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function, with three dimensions used for real space and three dimensions for phasespace, so that even if a modest number of cells is used per dimension the grid ends up being prohibitively large. Instead, one can expand the distribution function in phasespace to spherical harmonics and express it using the complex amplitudes of the expansion and a 1D grid for the magnitude of momentum [4].

$$f(\mathbf{p}, \mathbf{r}, t) = \sum_{l=0}^{\infty} \sum_{m=-l}^l f_l^m(\mathbf{p}, \mathbf{r}, t) P_l^m(\cos \vartheta) e^{im\varphi}$$

$$0 \leq \vartheta = \arccos(p_x/|\mathbf{p}|) < \pi, \quad 0 \leq \varphi = \arctan_2(p_y, p_z) + \pi < 2\pi$$

where $f_l^m = [f_l^m]^*$ the amplitudes of the expansion and P_l^m the associated Legendre functions. The spherical harmonics are characterized by two indices l, m , where large l, m correspond to highly directional/anisotropic harmonics (Fig. 1).

The main advantage of this approach is that angular scattering due to collisions between electrons and ions tends to isotropize the distribution function by rapidly damping the high-order harmonics. It is therefore possible to capture the physics using only few terms in the expansion, and reduce the problem size, compared to a regular Vlasov code, by a factor in excess of 1,000. Other advantages of this expansion are that it makes it easier to stretch the phasespace, that it allows one to resolve the magnitude of momentum much more finely than the angle, and that it makes it easier to include collisions between electrons. On the other hand, its main vulnerability, the singularity at the origin $p = 0$, can be effectively addressed in the numerical scheme.

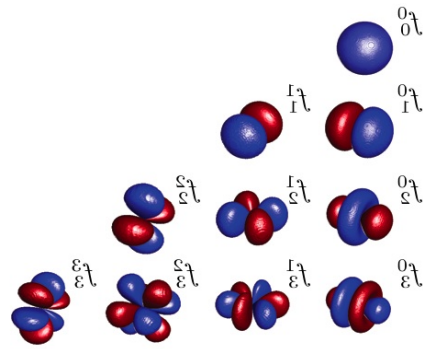


Figure 1: The first ten spherical harmonics

Numerical Grid

The expansion to spherical harmonics results in an irregular grid in phasespace (Fig. 2a), with the smallest cell at the origin. Numerical stability—assuming an explicit scheme—requires that a sufficiently small time-step and/or a high-order integration method be used to resolve the highest order harmonic at the smallest cell (e.g. Fig. 2b-2c). Alternatively one can restrict the number of harmonics resolved for low-momentum cells (Fig. 2d). This makes the code stable even for relatively large time-steps, but can lead to some artificial noise. At low-momentum cells, even a very small amount of collisionality damps this artificial noise immediately.

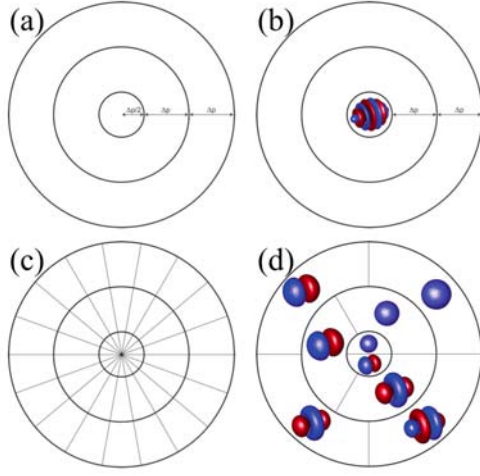


Figure 2: (a) First three $|p|$ -cells, (b)-(c) keeping all f_l^m for all $|p|$ -cells, (d) keeping only low f_l^m for low $|p|$ -cells.

Parallelization is achieved by decomposing the computational domain in real space, which is represented in a Cartesian grid.

2D3P Vlasov code

The following normalizations are adopted to model the Vlasov-Maxwell set of equations:

$$t \rightarrow \omega_p t \quad r \rightarrow k_p r \quad p \rightarrow p/(m_e c) \quad n \rightarrow n/n_p \quad q_e \rightarrow -1 \\ f \rightarrow c^3 f/n_p \quad \{E \text{ or } B\} \rightarrow e\{E \text{ or } B\}/(m_e c \omega_p)$$

where the subscript “p” indicates the maximum initial plasma density. The full 3D electromagnetic fields along with spatial advection in 2D (x,y) have been implemented in an explicit scheme following Ref. [4]. The object-oriented design allows for interchangeability of numerical operators, which has made it possible to incorporate a number of numerical methods and compare their performance in terms of speed, accuracy and stability. The code has been tested for standard plasma physics problems, e.g. the (collisionless) two-stream instability and (collisionless) plasma expansion. The evolution of these phenomena is modeled well up to a point, after which more harmonics have to be added to accurately describe the physics. Once collisions are added the need to continually add high order harmonics disappears. For collisionless plasmas the applicability of the expansion to spherical harmonics is limited to cases with slow spatial variation and weak electric field.

A laser source is mocked by substituting part of the distribution function with a new distribution function with a temperature determined by the characteristics of the laser. The code can generate output that includes the fields, the moments of the distribution function and allows for the conversion of the entire electron distribution function to a Cartesian grid so that it can be displayed using standard visualization software. Periodic and reflecting boundaries have been implemented.

Electron-ion collisions

Collisions involve predominantly low-energy particles. We assume that relativistic effects can be ignored in the collision operators and use the standard non-relativistic Fokker-Planck formalism in Ref. [5]. The expression for the evolution of the amplitude of each harmonic f_l^m due to collisions between electrons and ions becomes:

$$\left(\frac{\partial f_l^m}{\partial t} \right)_{ei} = -\frac{l(l+1)}{2} \nu_{ei} f_l^m$$

where $\nu_{ei} = n_i Y_{ei} / v^3$ with $Y = 4\pi(ZZ'e^2) \ln \Lambda / m^2$ and $Y_{ei} \equiv Y(Z'=1, m=m_e)$. Angular scattering leads to damping of each f_l^m with a rate proportional to $l(l+1)/2$. That is, the more

anisotropic/directional a harmonic is the faster it decays, as angular scattering tends to smooth out the shape of the distribution function. This makes it possible to truncate the expansion keeping only few harmonics for collisional plasmas. Additionally, because the collision-time scales with the cube of the velocity, even fewer terms are needed in the low-momentum cells. This relaxes the restrictions on the time-step significantly. To derive a characteristic time for angular scattering we use the expression for the RMS velocity $m\langle v^2 \rangle = 3kT$ to obtain

$$\tau_e = 3.44 \times 10^5 \times \frac{(T_e[\text{eV}])^{3/2}}{n[\text{cm}^{-3}] \times \ln \Lambda} \text{sec}.$$

The Coulomb Logarithm in the code is calculated using the expressions in the NRL Plasma Formulary.

Electron-electron collisions

The Fokker-Planck equation for any two species can be written as in Ref. [5]:

$$\frac{1}{Y} \left(\frac{\partial f}{\partial t} \right) = 4\pi \frac{m}{M} F f + \left(\frac{M-m}{M+m} \right) \nabla H(F) \cdot \nabla f + \frac{\nabla \nabla G(F) : \nabla \nabla f}{2}$$

where F is the distribution function and M is the mass of the scatterer and H, G (the Rosenbluth potentials [6]) are integral functions for F . This equation can be linearized by assuming that the anisotropic part of the distributions F, f is a perturbation to the isotropic part: $F = F_0^0 + F_a, f = f_0^0 + f_a$. This yields a nonlinear equation for the isotropic part and a linear equation for the anisotropic part of the distribution. The full derivation of the expressions for any two species will be presented elsewhere.

For collisions between electrons, where $F \equiv f$, the nonlinear equation for the isotropic part of the distribution can be written:

$$\left(\frac{\partial f_0^0}{\partial t} \right)_{ee} = \frac{4\pi Y_{ee}}{3} \frac{1}{v^2} \frac{\partial}{\partial v} \left[\frac{1}{v} \frac{\partial G}{\partial v} \right]$$

$$G = f_0^0 \int_0^v f_0^0 u^4 du + v^3 f_0^0 \int_v^\infty f_0^0 u du - 3 \int_v^\infty f_0^0 u du \int_0^v f_0^0 u^2 du$$

This formulation allows developing a numerical scheme that conserves energy and number of electrons. For velocities on the order of $\sqrt{kT/m}$ or larger any regular expression for the integrals above suffices. For $v \ll \sqrt{kT/m}$ the second and third terms in the equation for G vanish up to $O(v^5)$, and the Taylor-expanded form of f_0^0 must be used. This ensures that f_0^0 relaxes to a Maxwellian. An explicit scheme can be used for this nonlinear equation. The time-step for this integration can be a fraction of the time-step for the Vlasov part of the code, but since it involves only one harmonic f_0^0 and only a few simple calculations it does not affect the overall performance of the code.

To simplify the expression for high order collisions we define the integrals:

$$I_j(f_l^m) = \frac{4\pi}{v^j} \int_0^v f_l^m u^{j+2} du \quad J_j(f_l^m) = \frac{4\pi}{v^j} \int_v^\infty f_l^m u^{j+2} du$$

The equation for high-order harmonics becomes:

$$\frac{1}{Y_{ee}} \left(\frac{\partial f_l^m}{\partial t} \right)_{ee} = 8\pi f_0^0 f_l^m + \frac{I_2(f_0^0) + J_{-1}(f_0^0)}{3v} \frac{\partial^2 f_l^m}{\partial v^2} + \\ \frac{-I_2(f_0^0) + 2J_{-1}(f_0^0) + 3I_0(f_0^0)}{3v^2} \left(\frac{\partial f_l^m}{\partial v} - \frac{l(l+1)}{2} \frac{f_l^m}{v} \right) + \\ \frac{1}{2v} \frac{\partial^2 f_0^0}{\partial v^2} [C_4 I_{l+2}(f_l^m) + C_4 J_{l-1}(f_l^m) + C_2 I_l(f_l^m) + C_2 J_{l+1}(f_l^m)] + \\ \frac{1}{v^2} \frac{\partial f_0^0}{\partial v} [C_3 I_{l+2}(f_l^m) + C_4 J_{l-1}(f_l^m) + C_5 I_l(f_l^m) + C_6 J_{l+1}(f_l^m)]$$

This is a linear equation for f_l^m with the constants $C_i(l)$ given in the Appendix. $f_0^0(t + \Delta t)$, which has been calculated from the nonlinear equation above, can be used to integrate implicitly

for f_l^m . The first two lines in this equation yield the scattering of f_l^m due to the Rosenbluth potentials calculated from f_0^0 . This includes an “angular scattering” term $\propto [l(l+1)/2]f_l^m/v^3$. The last two lines can be seen as the effect that f_0^0 has on the Rosenbluth potentials calculated from f_l^m . We note that while the “scattering term” is of order $O(l^2)$, all other terms are of order $O(1)$ and they may be neglected for $l \gg 1$.

Benchmark

In order to study Shock Ignition the collision modules have been attached to the Vlasov code above. To test their accuracy we calculated the Spitzer heat conduction:

$$\frac{\mathbf{q}}{nm_e v_t^3} = - \left(\frac{320}{3\pi} \delta_T \epsilon \right) \frac{v_t}{\langle v_{ei} \rangle} \frac{\nabla T}{T}$$

where δ_T, ϵ are given in Ref. [7] and $\langle v_{ei} \rangle = Z/(\omega_p \tau_e)$. This includes the effects of both electron-ion and electron-electron collisions. We simulated a plasma with density $n=10^{23}/\text{cc}$ and a sinusoidal temperature profile with $T_{\min}=300\text{eV}$ and $T_{\max}=323\text{eV}$. Spitzer heat conduction was recovered with error less than 0.2% throughout the simulation box (with length 1000 mean free paths). If the Rosenbluth potentials calculated from f_l^m are neglected the error increases to 4%.

Appendix

Coefficients in the linearized Fokker-Planck:

$$C_1 = \frac{(l+1)(l+2)}{(2l+1)(2l+3)}; \quad C_2 = -\frac{(l-1)l}{(2l+1)(2l-1)}; \quad C_3 = \frac{-l(l+1)/2 - (l+1)}{(2l+1)(2l+3)};$$

$$C_4 = \frac{-l(l+1)/2 + (l+2)}{(2l+1)(2l+3)}; \quad C_5 = \frac{l(l+1)/2 + (l-1)}{(2l+1)(2l-1)}; \quad C_6 = \frac{l(l+1)/2 - l}{(2l+1)(2l-1)}$$

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Multi-component effects on the x-ray scattering signal from warm dense matter

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Introduction

High-power and high-energy laser sciences open the way to create and study warm dense matter (WDM) in a controlled laboratory environment. Characterized by near solid density and temperature of a few electronvolts, WDM contains the states between condensed matter physics and the ideal plasma regime. At these conditions, one has to describe matter in a highly correlated state that is also governed by electron degeneracy and bound states. Therefore, WDM still poses many challenges for both theoretical and experimental studies. From the theoretical point of view, the occurrence of strongly coupled ions prohibits the use of well-known perturbation methods developed for ideal plasmas whereas the degenerate electrons require a fully quantum-mechanical description.

WDM research is also driven by the aim to achieve energy by means of inertial confinement fusion [1, 2], where it occurs in the converters and as a transient state in the core. In addition, the properties of WDM are required to develop a better understanding of astrophysical objects as it naturally occurs in the interior of giant planets and in the outer layers of old stars such as white and brown dwarfs or neutron stars [3,4]. During the last decade WDM can be created in a rapidly increasing number of powerful laser facilities worldwide. First experiments on different materials, e.g., Be, Li, CH and LiH [5,6,7,8], have demonstrated that high energy density matter has very interesting properties.

Besides generation, the diagnostics of WDM poses a severe challenge. Due to the high particle density, energetic particle beams or x-rays are required to penetrate the system. The scattering of x-rays has been shown to reflect the microscopic structure as well as being able to determine the basic plasma parameters like electron density, ion charge and temperature [9]. The application of x-ray scattering as a diagnostic method however relies on theoretical models for the structure of the material under investigation since the plasma parameters are deduced as a fitting parameter that allow to match the scattered signal with the theoretical description.

For composite materials like LiH or CH multiple components must be included in the description of WDM. The mutual interplay between the different highly correlated ions constitutes additional problems for a consistent theoretical description of the x-ray scattering signal. In this contribution, we present a theoretical model to describe the elastic Rayleigh peak in a multi-component system. The structural information needed are obtained by a quasi-classical approach that uses the hypernetted chain (HNC) equations which can be extended to treat multiple ion species [11,12]. For classical ion-systems, HNC has been proven to describe strong coupling effects very well [10]. The degenerate electrons in WDM can only be incorporated in an approximate way: here, we use a linearly screened Coulomb potential for the ionic interactions which is screened by a Thomas-Fermi like screening length. Nevertheless, this approach provides an efficient method to analyse experimental data. In contrast, full quantum simulations such as density function molecular dynamics (DFT-MD) [12] are capable to fully describe a complex WDM system, i.e., strongly coupled ions combined with degenerate electrons.

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However, such a treatment requires a high numerical effort and its applicability as an experimental analysis tool is thus very limited. We mainly use it to benchmark the classical HNC calculations and to determine charge states and inter-ionic potentials [12].

Theoretical description of the scattering signal

The spectrum of the scattered radiation per solid angle $d\Omega$ and per frequency interval $d\omega$ is directly proportional to the microscopic structure of the electrons. In a many particle system this is expressed by the total dynamic electronic structure factor

$$P_s(k, \omega) d\omega d\Omega \propto S_{ee}^{tot}(k, \omega). \quad (1)$$

Here, k and ω denote the momentum and the energy change of the scattered photon, respectively. The structure factor for partially ionised matter, which is the Fourier transform of the correlation function of electron density fluctuations, can be decomposed with respect to electron fluctuations due to the ions and the free gas response (neglecting core excitations) [13,14]

$$S_{ee}(k, \omega) = |f(k) + q(k)|^2 S_{ii}(k, \omega) + Z S_{ee}^0(k, \omega). \quad (2)$$

The first contribution dominates the low frequency domain of the x-ray Thomson scattering signal and is strongly modulated by the strongly correlated ions. Here, electrons bound to the nucleus, characterised by the atomic/ionic form factor $f(k)$, and the electrons in the screening cloud, described by $q(k)$, contribute to the scattering of the probe beam. The second part of Eq. (2) describes the scattering due to the free electrons where Z denotes the number of free electrons per atom. This part describes the large frequency regime where the Compton shifted electron features as well as collective excitations (plasmons) can be observed. Effects of internal excitations and ionisation induced by the probe beam can be neglected in the low- Z materials considered here.

For the theoretical description of x-ray Thomson scattering, the microscopic structure expressed by the different structure factors has to be determined. The large frequency electron feature corresponds to the electron-electron structure factor of free electrons that is directly related to the correlation function of the density fluctuation by the fluctuation dissipation theorem [15]. In WDM, the electrons are degenerate and mostly weakly coupled. Thus, the random phase approximation (RPA) is an accurate description which can be improved by inclusion of electron-ion collisions (see e.g. Ref. [16]).

The ion feature, that describes the electrons co-moving with the ions, is directly related to the ion-ion structure factor describing the spatial arrangement and the thermal motions of the ions. Due to the finite bandwidth of the x-ray probe beam as well as the finite resolution of the detector, this quantity cannot be resolved in experiments with laser-driven sources. Thus, it is sufficient to use the frequency-integrated or static description $S_{ii}(k) = \int S_{ii}(k, \omega) d\omega$ for the ionic correlations. Here, the static structure factor is calculated using the HNC approach. As effective interaction potential between the ions, a Coulomb

potential linearly screened by the electrons is applied. This model yields similar results as *ab initio* simulations [12]. The atomic form factor of ions in plasmas can be approximated very well by the one for isolated atoms [17]. For light elements like beryllium where only 1s or 2s electrons occur, it is sufficient to use hydrogen-like wave function with an effective core charge [18]. The screening function $q(k)$ that specifies the (free) valence electrons surrounding the ions, can be approximated within the linear response theory, that is by

$q(k) = Z\kappa_e^2 / (\kappa_e^2 + k^2)$, where κ_e is the inverse electron screening length [19].

Recent experiments were performed on systems with more than one ion species [7,8]. This raises the question how the mutual correlations within the ion system affect the scattering process. We generalised Eq. (2) for systems with an arbitrary number of ion species using a derivation similar to the original Chihara approach for single ion species which splits the electron density into bound and free contributions [13,14]. The result for the elastic ion feature $W_R(k)$, that is the Rayleigh peak, can be summarised as

$$W_R^{\alpha\beta}(k) = \sum_{\alpha,\beta} \frac{\sqrt{n_\alpha n_\beta}}{n_i} (f_\alpha + q_\alpha)(f_\beta + q_\beta) S_{\alpha\beta}(k). \quad (3)$$

Here, the summation runs over the different ion species α and β with the corresponding ion densities n_α . n_i denotes the total ion density in the system. Moreover, the partial ion-ion structure factors need now to be considered. The HNC approach, which can be applied to systems with multiple ion species [11], naturally describes the mutual correlations between the different ion species. Similarly, *ab initio* calculation like DFT-MD can be used for systems with more than one ion species. However, the already high computational demand strongly increases here with the number of species. The determination of the atomic or ionic form factors as well as the screening functions for the various ion species can be done in a similar fashion as for simple one-component systems.

Results and Discussion

Figure 1 presents the ionic structure as well as the weight of the Rayleigh peak for a dense CH plasma. At the temperature considered, the ionisation state of the carbon ions is $Z = 2$ whereas hydrogen is fully ionised. In the lower panel of the figure, the partial structure factors are shown. Under these plasma conditions, the higher charged carbon ions exhibit a more distinct structure than the protons or the cross term. In fact, the protons are only moderately coupled with a classical coupling parameter of $\Gamma = 1.3$. They can be nearly described as a uniform background as demonstrated by the almost constant structure factor that is typical for a weakly coupled system. However, strongly coupled ion species can imprint their structure on the other ion species. Moreover, the mutual correlations in the multi-component system lead to an additional screening due to the other species that is self-consistently included within the HNC equations. Note, that the partial structure factor $S_{CH}(k)$ presents the typical characteristics of the cross terms where unity is not added in the definition [20] and negative values appear for small k (the long-range limit is zero). The total structure factor is however positive definite.

The upper panel of Fig. 1 shows the elastic ion feature (strength of the Rayleigh peak) for CH given by Eq. (3). The required form factor for carbon is taken for isolated double charged carbon [18]. Due to the fact that hydrogen is fully ionised, only the screening function and no atomic form factor has to be considered. This quantity is calculated in linear response as defined above (however, different screening clouds for carbon ions and protons are considered). The resulting weight of the Rayleigh peak is shown as a solid red line in Fig. 1. Furthermore, the different contributions related to the different

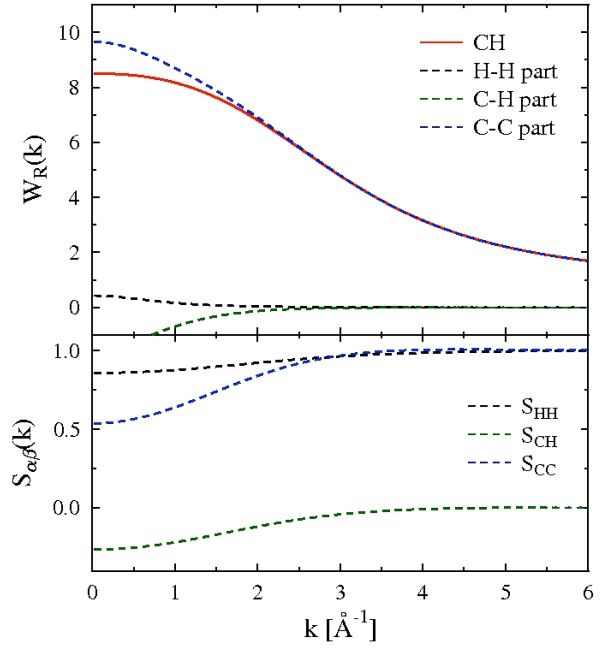


Fig. 1: The structure of a CH plasma with an ion density of $n_c = n_H = 5 \cdot 10^{22} \text{ cm}^{-3}$, a temperature of $T = 8 \text{ eV}$ and $Z_H = 1$ and $Z_C = 2$. The upper panel shows the weight of the Rayleigh peak (solid line) and its three contributions due to variations of carbon ions and protons (dashed lines). The lower panel shows the partial structure factors for CH.

combinations of C and H ions are plotted in the figure. This comparison makes it clearly visible that the elastic ion peak is mainly given by the scattering on the C^{2+} ions. For small k values the combination of all quantities are relevant whereas for larger distance, only the form factor of the carbon ions characterises the ion peak. Thus, forward scattering, which highlights the small k regime, is more sensitive to multi-component effects described here.

As a next example, we compare the strength of the elastic scattering for pure carbon, CH and a CH_2 with a constant carbon density to study the influence of the protons in the systems. In Fig. 2, the carbon structure factors are presented for

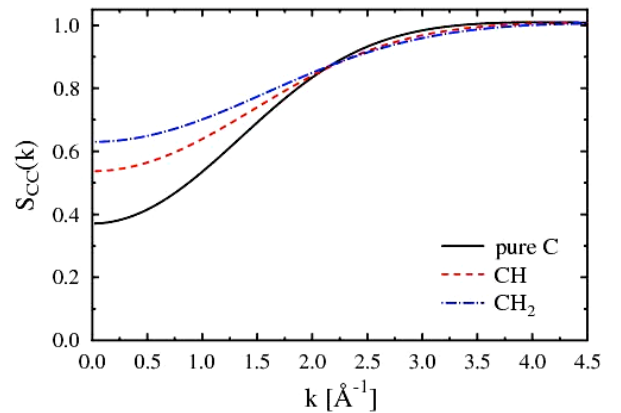


Fig. 2: Static carbon-carbon structure factors for warm dense carbon, CH and CH_2 calculated with the HNC method. The carbon density of $n_c = 5 \cdot 10^{22} \text{ cm}^{-3}$, the temperature of $T = 8 \text{ eV}$ and a charge state of $Z_C = 2$ are fixed for all systems. For the calculation of CH and CH_2 , fully ionised hydrogen, i.e. $Z_H = 1$, with densities of $n_H = 5 \cdot 10^{22} \text{ cm}^{-3}$ and $n_H = 10^{23} \text{ cm}^{-3}$ is applied, respectively.

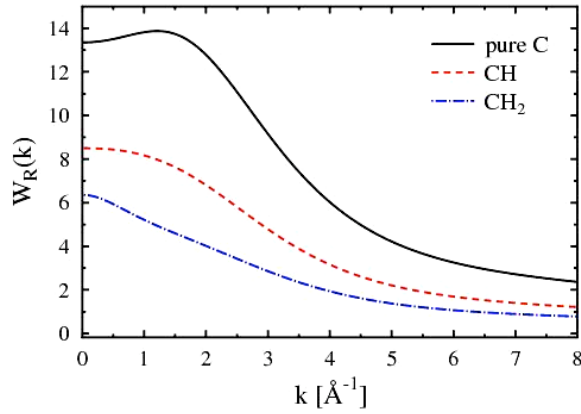


Fig. 3: Comparison of the weight of the Rayleigh peak for warm dense carbon, CH and CH₂. The plasma conditions are the same as in Fig. 2.

the different cases using one and two-component HNC with a screened Coulomb interactions, respectively. The pure carbon plasma shows the most pronounced structure. With the increase of proton density, the structure factor rises for small k values. This is due to the fact that the protons in the system contribute to further screening of the carbon-carbon interactions and, thus, the coupling decreases. Obviously, this effect becomes more significant with a higher proton density in the case of CH₂.

Fig. 3 shows the weight of the Rayleigh peak for the pure carbon, CH and CH₂ as in Fig. 2. The form factors for the carbon ions and the screening functions are calculated in the same manner as for CH described in Fig. 1. The weight of the Rayleigh peak for a pure carbon plasma simplifies to $W_R(k) = (f_C + q_C)^2 S_{CC}(k)$, whereas three contributions have to be added for the two systems of plastics. With the occurrence of protons in the system, the ion peak decreases nearly by a factor of two. This reflects the change in the partial structure factors due to the multi-component description. Furthermore, even though the weight of the Rayleigh peak is mainly dominated by the scattering on the carbon ions the fully multi-component description is necessary to get the correct statistical weight given by the densities of the elements considered. An increase of the proton density, like given with CH₂, leads to a further decrease of the elastic ion feature.

Conclusions

In this contribution, we investigate the effects of multiple ion species on the x-ray scattering process from warm dense matter. In particular, we discuss elastic scattering, that is, the weight of the Rayleigh peak. Based on partial structure factors from hypernetted chain solutions, a generalised approach of the theoretical model is applied to account for multi-component effects. We demonstrate that mutual correlations significantly influence the partial structure factors due to the fact that the ions with the highest charge imprint their structure on the other components. The weight of the Rayleigh peak that is directly related to the static structure factors is thus also sensitive to the interplay between the different correlated ions in the systems. These effects are especially pronounced in the case of forward scattering, i.e. for small k values. Furthermore, we demonstrate that the full multi-component description is also necessary for cases where x-ray scattering is dominated by one species. This effect is related to the differences in the statistical weight of the different contributions to the Rayleigh peak.

Acknowledgements

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Quasi-classical model of non-destructive wavepacket manipulation by intense few-cycle nonresonant laser pulses

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Introduction

This article follows a recent publication [1]. To quantify the observations of femtochemistry and coherent control, novel theoretical treatments were developed to accurately quantify the evolution of nuclear wavepackets created and controlled by the light-matter interaction. At low intensities (generally $I < 10^{10}$ Wcm⁻²), the evolution of a nuclear wavepacket can be described by the perturbation theory and the population of the initial state is assumed to be constant [2].

In the intermediate intensity regime (defined as 10^{10} Wcm⁻² < $I < 10^{12}$ Wcm⁻²), the population of the initial and final states must be considered; the intensity regime is also dependent on the coupling strength between the electronic states. The analytical solution of the Schrödinger equation is often impossible, hence numerical integrations or alternative representations of wavepackets are required: quasi-classical trajectories are often employed, which may be intuitively understood [3].

In the strong-field regime, which we define as $I > 10^{13}$ Wcm⁻², multiphoton or tunnel ionization become highly probable processes in atoms, and photodissociation and Coulomb explosion become dominant in molecules. Studying these processes has led to a significant advancement in understanding the dynamic structure of small molecules in intense laser fields, as recently reviewed by Posthumus [4]. While the interference of quantum pathways occurs under such conditions, it is often more useful and intuitive to think in terms of laser-induced dipole forces. The electric field induced by the laser field is sufficiently strong that atomic and molecular orbitals are polarized and impulsive Raman processes are possible [5]. The propagation of liberated electrons in the laser field should also be considered, particularly in the case of high harmonic generation (see [6] and references therein).

Under strong-field conditions it might seem unlikely that the delicate finesse required for quantum control would be possible; however, by applying the control pulse for a time significantly shorter than the fastest vibrational or rotational motions, the electronic structure of the molecule can be modified. This alters the environment in which the nuclei move, changing the motion of the nuclear wavepacket. An additional advantage is that ionization and fragmentation become less likely with shorter control pulses.

With the advent of few-cycle near infrared (NIR) laser pulses, a number of experimental groups have carried out studies to characterize vibrational wavepackets in hydrogenic molecular ions as recently reviewed by the authors [7]. Following illumination by an ultrafast strong-field pump, tunnel ionization of neutral H₂ or D₂ molecules leads to the formation of molecular ions supporting vibrational wavepackets. Such targets are attractive as they are theoretically tractable, and hence the Schrödinger equation can be solved within the Born-Oppenheimer and dipole approximations. Furthermore, the

excited electronic states are isolated from the ground state of the D₂⁺ ion, and hence electronic excitation is negligible. As is often true of vibrational wavepackets in diatomic molecules, the anharmonicity of the potential energy surface causes the wavepacket to dephase, dissipating the vibrational state components over a range of internuclear separations within a few classical periods. Theoretical predictions were recently confirmed experimentally, as reviewed by Calvert et al [7].

Just as femtochemistry led to quantum or coherent control, the observation of nuclear dynamics in strong-field few-cycle pulses also opens possibilities for analogous manipulation of the quantum state of molecules. Niikura *et al* have employed NIR laser pulses to perform experimental investigations of the laser-induced dipole forces to influence vibrational motion [8] in H₂⁺ and D₂⁺, focusing on measuring the ion fragmentation energy as the pump-control delay is varied. Thumm and co-worker have reported an accurate quantum mechanical treatment of a vibrational wavepacket [9] and the influence of an ultrashort control pulse [10]. The authors in collaboration with Murphy and McCann have used a similar numerical treatment to investigate the heating and cooling of vibrational populations [11], recently leading to the prediction of the 'quantum chessboard' effect [12].

The QCM is broken into a series of steps. The creation of a coherent superposition of states is modelled by extending the non-adiabatic ionization model of Yudin and Ivanov [13] to a molecular system, allowing the vibrational state population to be evaluated. A non-interacting classical ensemble is then created, with weighted quantized vibrational levels to reflect the initial state of the molecule. The wavepacket evolution is then approximated by allowing the classical ensemble to propagate on the internuclear potential energy surface (PES). Now, applying a 'control' pulse to the propagating ensemble causes a time-dependent deformation of the PES [8]: the large gradient of the induced electric field causes a large AC Stark shift. Essentially, the interaction of the induced field and the electron orbital causes a force on the nuclei. The resulting deformation of the potential accelerates or decelerates components of the ensemble depending on their direction of motion, transferring energy into or out of the system. The influence of this energy transfer is twofold, changing the relative phase of the ensemble components with respect to the unperturbed system, and transferring population between vibrational states. The transfer of population is considered to occur when the motion of an ensemble component is sufficiently perturbed by the control pulse that it takes on the characteristics of a higher- or lower-lying vibrational state (i.e. amplitude and frequency). Each of these processes is discussed below.

Molecular ionization in a few-cycle pulse

The MO-ADK (molecular orbital Ammosov-Delone-Krainov) formulation [14] has been relatively successful in predicting ionization rates; however, any theory that makes use of the original ADK treatment [15] is not ideally suited for a few-

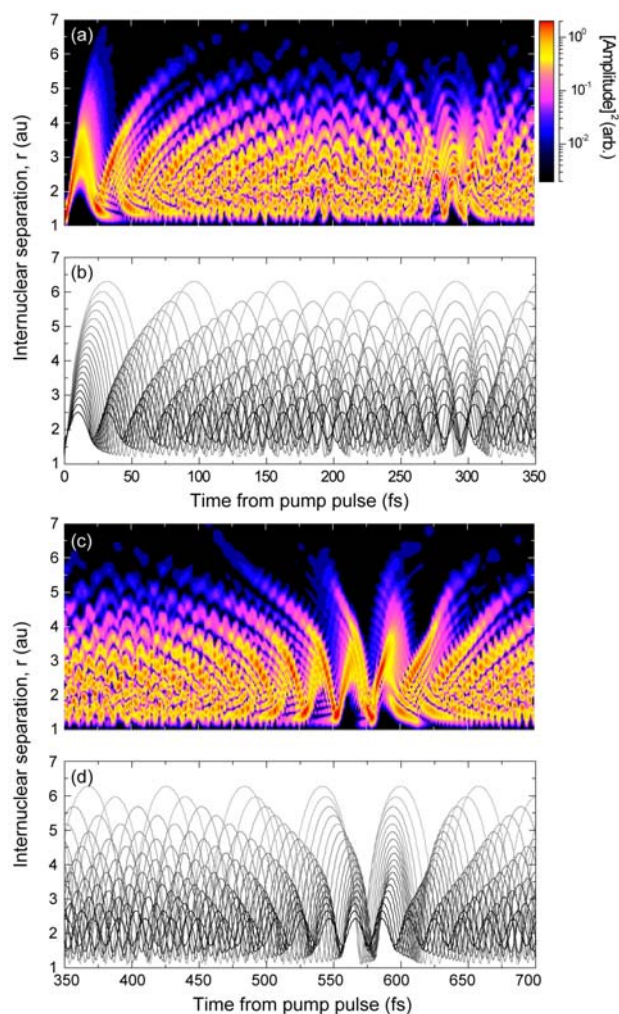


Figure 1. Comparison between the time-dependent solution of the Schrödinger equation for the evolution of the vibrational wavepacket ($v = 0-18$) in $D^+ 2$ (a, c, colour panels) and the QCM model (b, d, trajectories). The initial dephasing (a and b) and revival (c and d) of the vibrational wavepacket is shown. The QCM reproduces the features of the TDSE solution, as there is a bunching of the trajectories in regions where the solution of the TDSE is localized. The false colour panels are on a logarithmic scale, and the grey scale of the trajectories has been adjusted to illustrate similarities.

cycle pulse, being a quasistatic approximation. Instead, here we modify the recent work of Yudin and Ivanov [13] in which an analytical expression for ionization rate is found for arbitrary values of the Keldysh parameter. The flexibility of these expressions allows us to treat ionization between electronic states of the molecule efficiently.

The distribution of vibrational states depends on the probability of ionization and the overlap between the ground state wavefunction in the neutral and the available vibrational states in the ion. The recent experimental findings of Urbain *et al* [16] and theoretical discussion by Kjeldsen and Madsen [17] identified a significant deviation from the expected Franck-Condon-like distribution of vibrational states being populated by strong-field ionization in an ultrafast NIR laser pulse. This deviation was found to be dependent on peak laser intensity: on the leading edge of the pump pulse, the $D_2 \rightarrow D_2^+ (1s\sigma_g)$ ionization rate increases highly nonlinearly with laser intensity. The distribution of vibrational states is a direct result of the dependence of the ionization potential on internuclear separation, such that the maximum ionization rate (hence maximum population) does not necessarily occur at the optimum overlap of the ground state neutral wavepacket and the states in the molecular ion. Furthermore, once the intensity is

sufficiently high to generate the molecular ion, there is also a significant excitation rate to the $D_2^+ (2p\sigma_u)$ dissociative state. This is a loss channel and depopulates the number of D_2^+ ions supporting vibrational wavepackets. An additional loss channel exists in $D^+ + D^+$ Coulomb explosion, requiring a still higher intensity.

Field-free wavepacket evolution

In a manner directly comparable to the trajectory methods pioneered in femtochemistry and coherent control, we treat the evolution of the vibrational wavepacket in the molecular ion as an ensemble of classical particles moving on the bound PES.

The ionization event that launches the vibrational wavepacket enforces a coherence to the resulting motion, as the ionization rate is maximum for all vibrational states in the ion at the inner turning point. It is assumed that the unit mass of the classical particle is initially stationary following projection onto the ionic PES, which is reasonable considering that the tunnel time is much shorter than the characteristic time-scale for wavepacket propagation. We then treat the motion of each element of the ensemble as a Newtonian particle accelerated according to the differential of the PES. The presented discussion is confined to one dimension to be applicable to the hydrogenic molecular ion; however, this holds for any number of degrees of freedom, and hence has significant promise for quantifying wavepacket motion in complex polyatomics exposed to strong-field pump pulses causing excitation or ionization.

The motion of the ensemble (and hence the approximation to the wavepacket motion) is then predicted by allowing the unit masses to propagate as a function of time. The result of propagating the QCM for a typical number of vibrational states (generally from $v = 0$ to $v = 24$) is presented in figure 2, and a visual comparison made to the solution of the time-dependent Schrödinger equation (TDSE) for D_2^+ within the Born-Oppenheimer and dipole approximations, as published in [X]–[X]. Clearly, as the QCM generates a series of trajectories and the quantum treatment results in a continuous wavefunction, a direct numerical comparison is difficult. However, it is apparent from figure 2 that the QCM captures the essence of the quantum wavepacket result. This is not surprising, as both motions are the direct consequence of the PES shape as a function of R ; however, this comparison is intended to indicate the convergence of the classical trajectory calculation despite only requiring 7000 simulation steps to model the wavepacket motion over 700 fs. A typical split operator propagation of the TDSE (where a solution exists) requires of the order 30 000 steps, i.e. a time-step of 1 au (24.2 as).

Figure 1 presents two subsets of the now familiar wavepacket dephasing and revival. Comparing the QCM trajectories and the predicted TDSE-derived wavepacket amplitude at 590–600 fs, the intense features in the false colour representation of the wavepacket are apparent in the overlapping or closely spaced QCM trajectories. The slightly imperfect rephasing of the wavepacket predicted by the TDSE solution is replicated in the QCM trajectories.

Quantifying deformation of the PES

In the strong-field regime, the optically induced electric field is sufficient to not only distort the PES via the Stark shift, but also to polarize the molecular orbital(s), which the nuclei experiences as a time-dependent force, therefore changing the potential energy. The orbitals respond to the electric field (i.e. on an attosecond, or at least few femtosecond time-scale), but the nuclei are only driven by the cycle-averaged envelope; hence, if a control pulse is applied to a molecule supporting a vibrational wavepacket, the nuclear motion adjusts according to the external influence. Consequently, our control pulse will modify the effective PES by the dipole force generated by

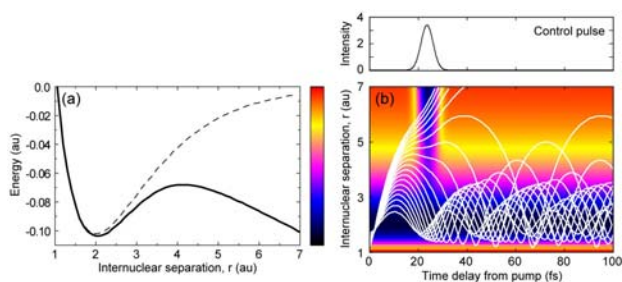


Figure 2. Potential energy surface distortion by the control pulse and associated ensemble perturbation. (a) With a control pulse intensity of $I_{\text{control}} = 3.5 \times 10^{13} \text{ Wcm}^{-2}$, the $1\sigma_g$ potential is heavily distorted (solid line) as compared to the field-free situation (dashed line). (b) The distortion of the PES as a function of time when a 7 fs control pulse is applied at 24 fs, referenced to the energy scale of (a). The trajectories predicted by the QCM are overlaid on the potential; the distortion of all trajectories is dependent on state amplitude and phase relative to the control pulse (shown in units of 10^{13} Wcm^{-2}). The maximum in the PES induced by the control pulse around 4 au present in (a) becomes a saddle point as a function of time in (b).

polarizing the molecular orbital and subsequent Stark shifting.

In figure 2(a), the field-free PES and the maximally laser-distorted PES are shown; such a dramatic deformation of the PES is only present for a fraction of the vibrational period. The distortion of the PES by the control pulse causes a time-dependent variation of $dV(R)/dR$, and hence the ensemble components experience an additional acceleration, the direction and magnitude of which depends on the direction the component is moving and its location on the PES. This causes a net increase or decrease of energy depending on whether the control pulse dipole force acts to enhance or retard the ensemble elements, interpreted as a change of vibrational state in a quantum representation. This is illustrated in figure 2(b), whereby a 7 fs FWHM Gaussian control pulse of peak intensity $I_{\text{control}} = 3.5 \times 10^{13} \text{ Wcm}^{-2}$ is applied 24 fs after the wavepacket is created by the pump pulse (i.e. $t = 0$). The trajectories around $R = 5 \text{ au}$ at 24 fs illustrate this point well: both are accelerated to larger R under the influence of the control.

Variation of vibrational state and phase

As the ensemble of quasi-classical particles propagates, the laser-induced force transfers energy into or out of the system. Clearly, the discrete vibrational states have an influence when considering the quantum nature: a change of vibrational state only occurs when sufficient energy has been gained or lost to instigate a change in state.

As is illustrated in figure 2, the time-varying distortion of the molecular PES by the control pulse causes the component trajectories to vary from the unperturbed motion. If additional energy is transported into a trajectory, the amplitude of the oscillation increases, along with the vibrational period, and hence the trajectory takes on the characteristics of a higher-lying state. We propose that this increase in energy is equivalent to population being transferred from a lower to higher vibrational state. Similarly, the control pulse can simultaneously remove energy from the system, causing a decrease in vibrational state.

As the control pulse can up- and down-shift vibrational population, it is possible that multiple ensemble components can end up in the same vibrational state, and the phase and relative population of each state must be considered. Variations in the phase and population are quantified by making a numerical comparison between the perturbed (e.g. figure 2) and unperturbed (figure 1) trajectories. Importantly, this comparison must be made once the intensity of the control pulse has dropped to zero, such that the trajectories are only defined by

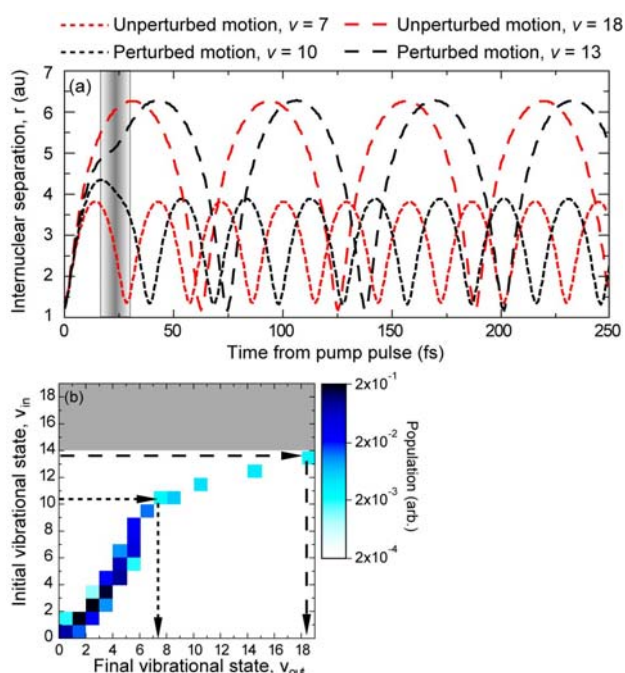


Figure 3. Vibrational population transfer in the QCM. (a) The application of a 7 fs duration control pulse with an intensity of $3.5 \times 10^{13} \text{ Wcm}^{-2}$ at 24 fs causes both positive- and negative-going population transfer. The initial state $v = 10$ is down-shifted into resonance with the $v = 7$ state by the control pulse (red and black short dashed lines), while the $v = 13$ state is up-shifted into resonance with the $v = 18$ state (red and black long dashed lines). The shaded region indicates the intensity of the control pulse and the vertical lines indicate when the intensity is $3.5 \times 10^{12} \text{ Wcm}^{-2}$. (b) The vibrational population matrix conveniently illustrates the action of the control pulse. Each point on the matrix indicates the likelihood of population transfer on a logarithmic colour scale, and the grey region for $v > 14$ indicates states are no longer bound. Projecting the sum of the matrix vertically returns the final vibrational population.

the static PES. In practice, it is important to ensure that the control pulse(s) does not suffer from significant pedestals on the femtosecond and picosecond time-scale. Therefore a third-order autocorrelation contrast measurement is required when comparing with experimental results, in case an underlying PES distortion exists.

Comparison with established results

While figure 1 has demonstrated that the QCM can successfully reproduce the unperturbed motion of a vibrational wavepacket by propagating the ensemble, it could be argued that any treatment relying on the differentiation of the field-free PES will give a similar result, assuming sufficient numerical accuracy. It is therefore vital to quantify the ability of the QCM to accurately describe the action of the control pulse; to this end, we compare the output of the QCM to established theoretical results. In figure 4, systematic scans of the control delay and intensity with respect to the pump pulse are presented for direct comparison with the reproduced results of Niederhausen and Thumm, figure 5 [10]. In both cases, the control pulse is 6 fs in duration and the initial vibrational state distribution is calculated using the tunneling theory. Importantly, the initial distribution of vibrational states in [10] was calculated using the ADK treatment [15]. The TDSE was then solved within the Born–Oppenheimer approximation, and the control pulse causes Raman transitions between vibrational states. Furthermore, following the control pulse, the wavepacket is projected onto the Coulomb explosion PES.

Despite the differences in method, a good agreement is found (especially considering how disparate the two numerical techniques are), indicating that the QCM accurately captures the

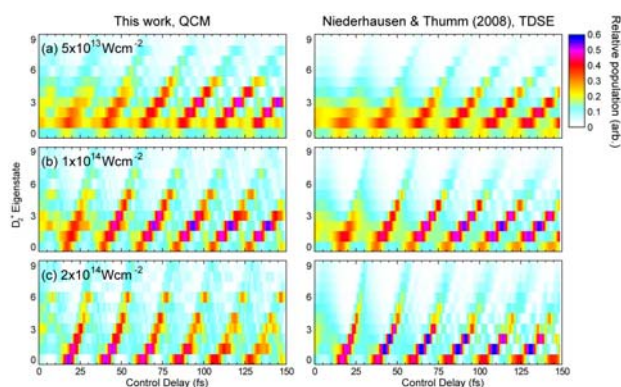


Figure 4. Final vibrational state population distributions for three different control pulse intensities as the temporal separation between the pump and control pulse is varied. The results of Niederhausen and Thumm [10] are reproduced for comparison with the QCM, which reproduces the periodicity and relative shift of vibrational population as the delay between the pump (6 fs, 10^{14} Wcm $^{-2}$) and the control is varied.

modification of the vibrational population as revealed by a full quantum mechanical model. Approximating the evolution of a wavepacket under the influence of an ultrafast strong-field control pulse as a quasi-classical ensemble is therefore justified.

Conclusions

A QCM has been proposed that allows the quantification of wavepacket dynamics modified by an ultrashort strong-field non-resonant laser pulse. We have discussed how the vibrational phase and population are adjusted by the control pulse, and a comparison has been made with established theoretical predictions. Systematic predictions of wavepacket dynamics as a function of pump intensity and control delay and intensity have been presented. Such results will be of interest to groups attempting to experimentally detect the manipulation of a wavepacket. Attosecond studies in complex many-electron molecules is such an application; the QCM could be employed to identify the populated electronic states suggested, whereby the coupling of electronic states in a strong field distorts the vibrational wavepackets generated, thus revealing the nuclear and electronic dynamics.

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Monochromatic XUV-photon + strong-field NIR cross-correlation by atomic excitation and ionization

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Introduction

Electronic transitions in matter are responsible for dynamic processes from atomic to macroscopic scales, including the optical properties of materials, chemical bonding and reactivity, conduction, the photoelectric effect and photosynthesis. While electronic transitions can be initiated collisionally, modern physics and chemistry often relies on the absorption of one or more photons over femtosecond to attosecond timescales (10^{-15} to 10^{-18} seconds) to observe such processes. The simplest case is when a single photon is absorbed resonantly. As the photon energy is increased, more tightly bound electrons are accessed, allowing atomic core states to be probed; perturbation theory is generally sufficient to describe such processes. Increasing the photon flux makes the simultaneous absorption of more than one photon likely, resulting in highly nonlinear nonresonant processes which are often described in terms of electronic distortions by the laser-induced electric field; this is generally referred to as the strong-field regime corresponding to an intensity of 10^{13} Wcm⁻² or higher.

Light source technology defines the parameters which may be explored and controlled. In addition to photon energy and intensity (or flux), key characteristics are bandwidth, duration and repetition rate. Loosely, the photon energy defines the type of transition that can be accessed in a resonant regime or whether transition should be considered as multiphoton or tunnelling in strong-field regime.

Synchrotron radiation, first observed in 1945, is the result of an energetic electron being deflected by a magnetic field, releasing energy in the form of photons. The photon energy is widely tunable by controlling the electron energy, magnetic field strength and bend radius. From the 1960s onwards, massive amounts of information was learnt about electronic processes from tens of eV to hundreds of keV [1]; this continues in Diamond, which produces photons with a high flux and repetition rate, and broad energy tunability. However the photons have poor coherence and pulses tend to be limited to picosecond durations.

The advent of the laser in 1960 saw an alternative method for generating photons which are inherently highly coherent. Advances in technology led first to pulsed operation, followed by broadband amplification techniques that allowed the generation of infrared pulses only a few cycles in duration, hence on femtosecond timescales [2,3,4]. By carefully controlling the temporal and spectral phase of light generated by the oscillator and amplifier, pulses can approach the transform limit. Furthermore, the electric field of these laser pulses is now controllable such that the phase between pulse envelope and carrier oscillation are identical for subsequent pulses. Essentially it is possible to define the group and phase velocities across few hundreds of nanometers of bandwidth.

Amplifier technology now means it is possible to generate electric fields of several volts per Angstrom on femtosecond timescales [5]. Modern laser sources are therefore coherent, generate high intensities, are capable of generating ultrashort pulses, however tend not to be tunable and are limited to operate in the IR to UV.

Free-electron lasers (FELs) combine elements of synchrotron and optical laser technology. A relativistic electron beam is passed through a periodic magnetic field in an undulator causing the emission of synchrotron radiation. By employing a series of long undulators FELs can be made to operate in the VUV to x-ray regimes. Such XFELs produce radiation coherent with the electron beam oscillation, causing a temporal bunching and laser gain through self-amplified spontaneous emission (SASE). XFELs such as FLASH in Germany and the LCLS in the US generate a very high intensity over a photon energy range of 10 eV to 1 keV in a pulse tens of femtoseconds long. Noise inherent to the XFEL construction and the SASE process causes a temporal jitter comparable to the pulse duration. While varying the electron beam energy will in principle make the photon energy tunable, in practice XFELs tend to have limitations to their energy flexibility.

High harmonic generation (HHG) in an intense ultrafast laser pulse is an intriguing compromise between an optical and free-electron laser. Following tunnel ionization, the liberated electron propagates in the time-varying laser field, and depending on the instant of release with respect to the laser electric field, can be accelerated back towards the parent ion [6]. Just as with electron impact excitation and ionization experiments, there is a small but measurable likelihood of the electron recolliding with the ion, recombining to a bound state and emitting excess kinetic energy as a photon.

The photon energy, flux and duration emitted during HHG depend on the wavelength, intensity and duration of the optical drive laser field [6]. Furthermore it is vital to observe phase matching conditions between the focus of the laser pulse and the HHG target. Optimal harmonic efficiency is possible by carefully controlling the size of focal volume and number density of the target such that harmonic photons are emitted in as constructively as possible.

By making the drive laser pulse as short as possible [7], typically limited to a few optical cycles, HHG produces an essentially continuous XUV spectrum. By filtering out the lower harmonic orders and minimizing the spectral phase variation over the continuous region, attosecond duration XUV pulses have recently been demonstrated by a number of research groups around the world. Direct observations of electron dynamics are now possible [7]; the majority of experiments have focused on atomic targets. While temporally resolving the electronic motion is a breakthrough, attosecond

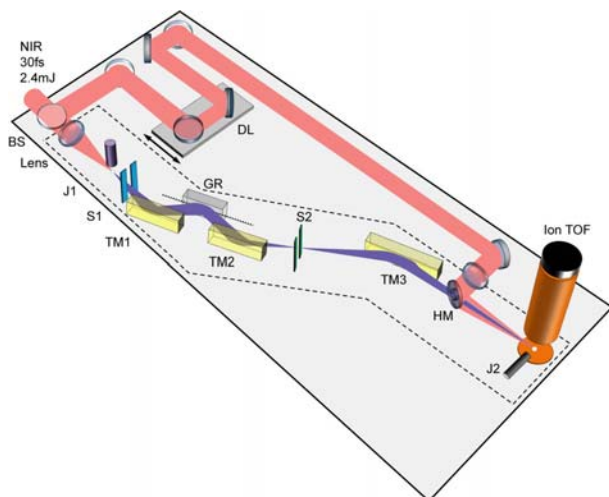


Figure 1. Schematic of XUV beamline including monochromator and NIR delay line. The dashed line indicates UHV chambers, BS – beamsplitter, J1 and J2 – effusive gas jets, TM1-TM3 – toroidal mirrors, GR – grating, S2 – slit, HM – annular mirror.

sources have the disadvantage of requiring a large bandwidth, typically tens of eV at a centre photon energy of ~ 80 eV, so will excite a broad range of electronic transitions.

HHG driven by an optical laser is the one element of the Artemis Facility and has been demonstrated to generate UV-XUV photons [8]. To make this source more flexible when studying electronic processes, it is now possible to select a single harmonic with an all-reflective monochromator designed and built at LUXOR [9,10,11]. This novel technique combines the femtosecond duration of optical lasers, an energy comparable to attosecond sources while maintaining the narrow bandwidth of XFEL sources, all with an inherent synchronization to a strong-field optical laser pulse. In this commissioning experiment, we intend to demonstrate and measure the temporal duration of the single-harmonic XUV beam by cross-correlating the XUV pulse with an NIR pulse focused to high intensity.

XUV-pump NIR-probe apparatus

The experimental apparatus employed in this work is schematically illustrated in figure 1; the majority of this section of the Artemis beamline has been discussed in recent Annual Report contributions. A 30 fs 4 mJ 800 nm pulse is split 3:1, the weaker beam is transmission focused into an argon gas jet (J1) to an intensity, $I > 10^{13}$ Wcm $^{-2}$, generating broadband UV-XUV photons up to ~ 100 eV by HHG [8]. An all-reflective monochromator optimized for the transmission of harmonics 13 to 29 (H13-H29, assuming H1 = 1.56 eV) is used to extract a single harmonic from the XUV region of the spectrum. This monochromator, designed and built by LUXOR, Padova [9,10,11], consists of a recollimating toroidal mirror, a grating illuminated parallel to the ruling direction, a second toroidal mirror followed finally by a variable width slit in the dispersion plane. By changing the angle of the grating with respect to the incoming XUV beam, the transmitted wavelength is varied and the bandwidth is controlled by changing the slit width.

Following the monochromator, the XUV beam is reflection focused into the interaction region of an ion time-of-flight mass spectrometer (TOFMS) [12]. The focus of the XUV beam is overlapped with a strong-field NIR pulse, generated by transmission focusing the stronger beam from the 3:1 splitter off an annular mirror. Clearly there is a significant energy loss on the annular mirror however the focus at the TOFMS is small enough to produce an intensity in excess of 10^{13} Wcm $^{-2}$. The relative path length of the NIR and XUV beams is variable

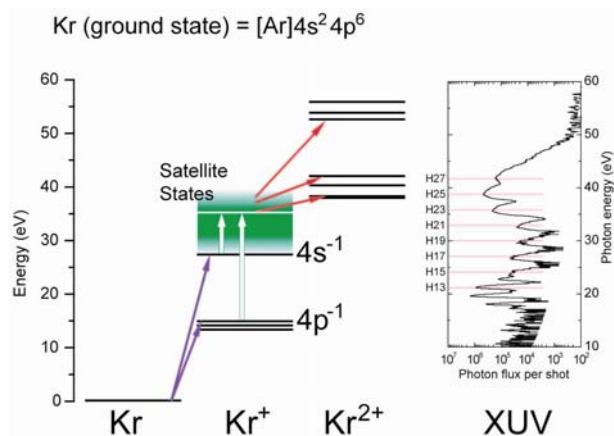


Figure 2. Energy level diagram for krypton showing relevant electronic transitions. The XUV pulse removes a 4s or 4p electron creating Kr $^+$ (violet arrows), populating highly excited satellite states through shake-up or the configuration interaction (green arrows). The NIR strong field pulse then tunnel ionizes the satellite states (red arrows). The XUV harmonic spectrum is shown (see figure 3).

using a translation stage, which introduce a NIR-XUV delay with a sub-cycle resolution and stability.

Krypton is effusively released into the interaction region of the TOFMS through jet J2. Over the H21-H27 XUV photon range (32.8 – 42.1 eV), krypton will absorb a single photon causing ionization of a 4s or 4p electron with cross sections of $10^{-1} - 10^{-2}$ or $10^0 - 10^1$ Mb (1 Mb = 10^{-22} m 2) respectively [13-15]. This leads to the population of highly excited satellite states through shake-up and the configuration interaction [16] as illustrated in figure 2. Now, assuming the NIR pulse arrives after the XUV pulse, the populated satellite states will be readily ionized with near-unity probability, generating Kr $^{2+}$. However if the NIR pulse precedes the XUV pulse, the NIR intensity is too low to generate significant amounts of Kr $^{2+}$, so will produce Kr $^+$ in low-lying electronic states with a probability per NIR pulse of the order 10^{-4} per shot. As a result, the likelihood of the subsequent absorption of an XUV photon is very low. Furthermore, while the XUV photon energy is sufficient to generate Kr $^{2+}$ directly through single-photon double ionization (apparent in figure 2), the cross section is extremely small and indeed only becomes observable with a photon energy in excess of 200 eV or a target pressure approaching 10^{-1} mbar [17].

Characterization of XUV HHG source

To measure the HHG spectrum, a calibrated channel-electron multiplier (CEM) is inserted into the XUV beam after the slit. By recording the CEM output current on an electrometer as a

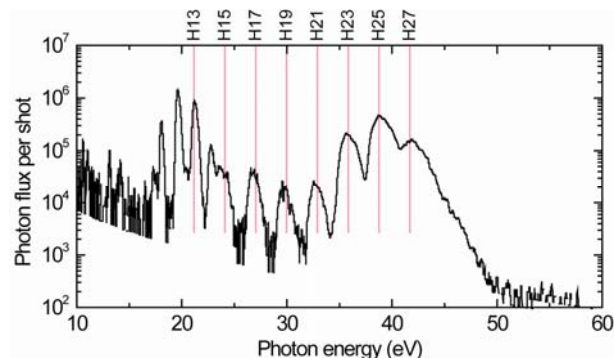


Figure 3. Monochromator transmission as a function of photon energy, corrected for CEM sensitivity (electrons per photon) and dispersion as a function of angle.

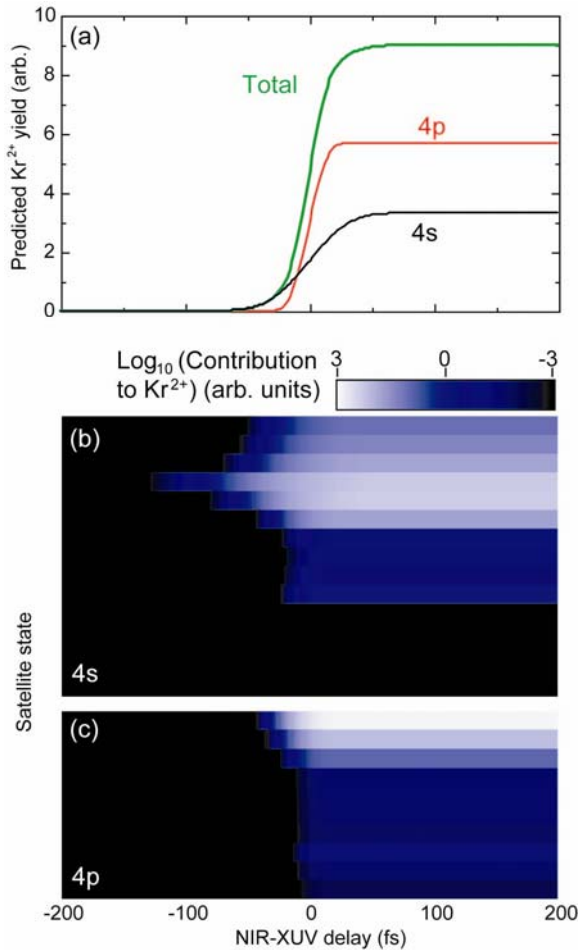


Figure 4. Predicted Kr^{2+} yield as a function of NIR-XUV delay. (a) Total yield along with contributions from ionization of satellite states populated by 4s and 4p ionization. (b) Contribution from individual 4s satellite states as identified in table 1, (c) contribution from 4p.

function of grating angle, the HHG spectrum following the monochromator is measured as presented in figure 3. A number of corrections are applied: the current is converted to number of photons and a Jacobian transform is applied for monochromator transmission as a function of grating angle. Also note the change in harmonic peak width with photon energy which is a consequence of angular dispersion being nonlinear with grating angle. In the present work, we selected a grating optimized to transmit most efficiently between 20–40 eV and at a high resolution, which in this case corresponds to a bandwidth of around 0.7 eV.

Modelling XUV+NIR yield

The duration of the HHG spectrum emitted in J2 (see figure 1), to a reasonable approximation depends on the ionization probability in the 30 fs drive pulse. For a $1.2 \times 10^{13} \text{ Wcm}^{-2}$, 800 nm 30 fs pulse, significant ionization is limited to within a FWHM of 14 fs. This will result in harmonic pulses of FWHM duration between 6 and 9 fs for H21–27. Clearly, the finite size of the illumination spot on the grating and the limited XUV bandwidth will temporally stretch the XUV pulse. To fully quantify this effect, we predicted the amount of Kr^{2+} enhancement depending on the relative delay between the XUV and NIR pulses as a function of XUV photon energy, performing a cross-correlation measurement on the atomic scale. We independently predict the population of the satellite states as a function of time through the XUV pulse according to the pulse intensity and cross section derived from synchrotron measurements [13–15]. Ionization of the satellite states by the strong-field is then modelled using the nonadiabatic theory of

4s satellite states		4p satellite states	
E (eV)	State	E (eV)	State
28.27	$4s^2 4p^4 (^3P) 5s$	30.65	$(^3P)5p(^4P_{3/2}) + (^4P_{5/2})$
28.70	$4s^2 4p^4 (^3P) 5s$	31.16	$(^3P)5p(^4D_{3/2})$
30.06	$(^3P)4d(^4D_{5/2})$	31.22	$(^3P)5p(^2P_{3/2})$
30.23	$(^3P)4d(^4P_{1/2}) + (^2P_{1/2})$	31.37	$(^3P)5p(^2P_{3/2})$
32.08	$(^1S)5s(^2S_{1/2})$	31.57	$(^3P)5p(^4D_{3/2})$
33.94	$(^1D)4d(^2S_{1/2})$	31.61	$(^3P)5p(^2D_{3/2})$
34.07	$(^3P)6s(^4P_{1/2}) + (^4P_{3/2})$	31.65	$(^3P)5p(^2S_{1/2})$
34.15	$(^3P)5d(^4D_{1/2})$	32.63	$(^1D)5p(^2P_{3/2})$
34.39	$(^1S)6s(^2S_{1/2})$	34.95	$(^1S)5p(^2P_{3/2})$
36.48	$(^1D)5d(^2S_{1/2})$	35.88	$(^1D)5d(^2G_{9/2})$
37.84	$(^1D)7d(^2S_{1/2})$		
38.99	$(^1D)8d(^2S_{1/2})$		
39.31	$(^1D)9d(^2S_{1/2})$		
39.47	$(^1D)10d(^2S_{3/2})$		

Table 1. 4s and 4p satellite state energies relevant to modelling XUV+NIR cross correlation. Taken from [16].

Yudin and Ivanov [18]. The delay between the XUV and NIR pulses then defines the population of the satellite states that are ionized to Kr^{2+} ; the free parameters are then the XUV pulse duration and the intensity of the NIR pulse.

While the cross sections of 4s and 4p ionization differ by at least an order of magnitude, Calo *et al* [16] have recently observed that the majority of the populated satellite states follow the photon energy dependence of the 4s or 4p cross sections. We assume their observations apply in the case of HHG XUV photon absorption. The predicted total Kr^{2+} yield is presented in figure 4(a) as a function of NIR-XUV delay for H25 and a NIR peak intensity of $1.2 \times 10^{13} \text{ Wcm}^{-2}$. The different rise times of the 4s and 4p contributions is a result of the relative populations of the satellite states as presented in figure 4(b) and (c), identified in table 1.

Kr^{2+} yield and pulse duration measurement

Before recording the ion yield, the zero delay point has to be identified which could be performed by recording a long delay scan and measuring the Kr^{2+} yield. A much faster alternative is rotating the monochromator grating to allow the zeroth diffraction order to propagate along the beamline; the zero order contains all harmonic orders along with the residual NIR drive radiation. The ZO NIR is synchronized with the XUV pulse, so by interfering the ZO NIR with the NIR reflected from the annular mirror on the exit of the TOFMS, the zero delay time is identified to within an optical cycle.

With the monochromator set to transmit one harmonic, room temperature krypton gas is then effusively released into the interaction region of the TOFMS to a local pressure of $\sim 10^{-4}$ mbar. Such a high pressure is required as the probability of ionization in either the XUV or NIR pulses is small. Ion time-of-flight spectra are averaged over 2500 laser shots as the NIR-XUV delay is scanned from -200 to 200 fs as presented in figure 5. A least squares fit between the experimentally observed and theoretically predicted Kr^{2+} yield is performed. Note that the model is integrated over the focal volume of the NIR pulse. The best fit is found with a H25 XUV pulse duration of 24 ± 2 fs which is in reasonable agreement with propagating a 8 fs XUV pulse through the monochromator.

Conclusions

By combining high harmonic XUV radiation with a strong-field NIR pulse, single-photon absorption and tunnel ionization have been combined to allow a cross-correlation measurement of the XUV pulse duration. This is made possible by the XUV pulse populating highly excited satellite states in Kr^+ which are subsequently ionized with a high probability by the NIR pulse. As the delay between the NIR and XUV pulses is scanned, the delay dependent satellite state population regulates the

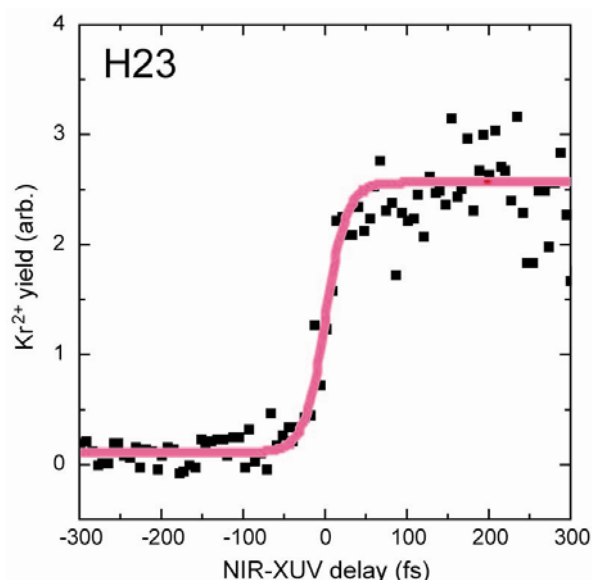


Figure 5. Measured and predicted Kr^{2+} yield as a function of NIR-XUV delay for harmonic 23 (photon energy 35.9 eV, wavelength 34.6 nm). The best least-squares fit reveals the rise of the yield “step” is the result of H23 having a duration of 24 ± 2 fs.

measured Kr^{2+} ion yield. This experiment demonstrates that gas-phase atomic and molecular physics can be performed with monochromated XUV radiation from a laser-driven gas source. This technique is highly complementary to HHG attosecond and femtosecond XFEL observations, with inherent sub-femtosecond synchronization to a strong-field NIR pulse and photon energy tunability. The photon flux is the main limitation to this type of measurement, and will be enhanced by at least an order of magnitude by the superposition of non-related frequencies [X] which has the added benefit of more closely spaced harmonic peaks.

Acknowledgements

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Multi-pulse scheme for controlling electron localisation upon molecular dissociation

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Introduction

Intense ultrashort pulses produced from titanium sapphire (Ti:sapph) laser systems provide powerful tools for probing and controlling molecular dynamics. With each technological development, new applications are found for these highly versatile systems. We propose a new application of this technology in quantum control of electron dynamics.

A significant development in this ultrashort pulse technology has arisen via spectral broadening techniques (such as self-phase modulation [1] and filamentation [2]), where it is now possible to generate pulses with durations on the order of a few cycles of the carrier field (where 1 cycle = 2.66 femtoseconds for 800 nm). High-impact applications of such pulses have been found in the production of XUV pulses via high harmonics [3, 4], the generation of attosecond pulses [5] and the probing of molecular motion [6-10].

For such short pulse durations, the position of the electric field oscillation relative to the pulse envelope becomes extremely important. This is appropriately quantified as the carrier-envelope-phase (CEP) offset, ϕ , as indicated in the pulse profile shown in Figure 1 (a). For ultrafast experimental applications, it is extremely advantageous to be able to fix this CEP, now possible through stabilisation loops [11].

An exciting application of CEP-fixed few-cycle pulses is in the quantum control of molecules. In this report we describe a new, multi-pulse scheme to achieve and enhance electron localisation upon dissociation, using bound vibrational wavepacket dynamics. Using recent theoretical results [12] we will show that this scheme not only utilises an alternative mechanism to other techniques [13, 14], but can also lead to significant enhancement in localisation via manipulation of the bound wavepacket.

Electron Localisation in D_2^+

The hydrogen molecular ion is an ideal candidate when studying fundamental dynamics in intense fields. As a three-body problem it is theoretically tractable, especially when treated within the Born-Oppenheimer and dipole approximations. The larger reduced mass of the deuterium molecular ion makes it more suitable for experimental studies of wavepacket dynamics with ultrashort laser pulses, having vibrational periods of the order of 20 fs.

When intense (up to 4×10^{14} Wcm⁻²) near-infrared laser pulses interact with the deuterium molecular ion, only two electronic states are significantly coupled and need to be considered when treating the resulting dynamics.

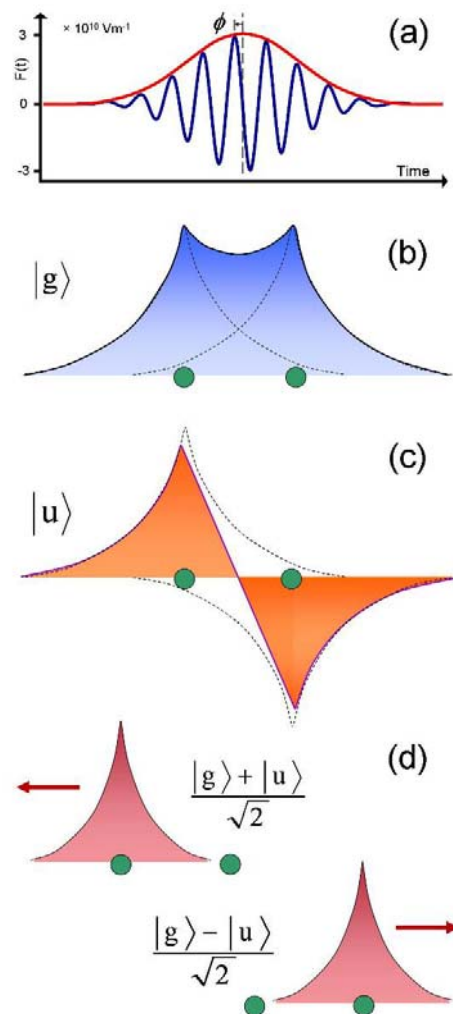


Figure 1. (a) Sketch of the Electric field for a CEP stabilised pulse where the CEP offset has been set to $\pi/2$. (b) and (c) Schematics of the electron orbitals for the two lowest lying states of D_2^+ , $1s\sigma_g$ ($|g\rangle$) and $2p\sigma_u$ ($|u\rangle$). (d) How electron localisation can be determined if dissociation occurs via a superposition of the two electronic states for a particular R value.

As sketched in figure 1 (b) the wavefunction of the lowest energetic state ($|g\rangle$) corresponds to a high probability of the electron being located between the two deuterons, creating a bound state. The second accessible state ($|u\rangle$) has an equal probability of the electron localising to either nucleus, and is a dissociative state. If dissociation occurs via one of these two

states, the electron has equal probability of being bound to one nucleus or the other.

However, if a nuclear wavepacket can be manipulated to dissociate via both these channels simultaneously, (i.e. in a superposition of the two electronic states) then it becomes possible to determine which nucleus the electron localises to; examples of this are shaded in red in figure 1 (d).

Figure 1 displays how the wavefunctions of the electronic states can be rewritten into a new basis set where it is possible to determine which of the nuclei the electron will localise to should photodissociation occur via both states. As described by Kling *et al.* [Kling] the new nuclear wavefunctions can be expressed as:

$$\begin{aligned}\phi_l(R,t) &= \frac{\phi_g(R,t) + \phi_u(R,t)}{\sqrt{2}} \\ \phi_r(R,t) &= \frac{\phi_g(R,t) - \phi_u(R,t)}{\sqrt{2}}\end{aligned}\quad [1]$$

Where ϕ_g and ϕ_u are the dissociating nuclear wavefunctions on the $1s\sigma_g$ and $2p\sigma_u$ states respectively. We can then define an absolute asymmetry parameter for the localisation as

$$A = \int |\phi_l(R,t)|^2 dR - \int |\phi_r(R,t)|^2 dR \quad [2]$$

It is worth noting that dissociation does not occur after every pulse interaction, and this point is implicit in the above definition. Elsewhere [13, 14] a normalised asymmetry parameter is used, where the asymmetry is expressed as a percentage of the probability of dissociation. However this can be misleading as it may infer a large asymmetry in cases where the actual probability of dissociation is low. In the work discussed here, we place an emphasis on achieving large asymmetry *and* significant dissociation probability.

The Multi-pulse Scheme

It follows from Equation (1), that the requirement for controlling electron localisation upon dissociation is to ensure that fragmentation of the molecule occurs in a superposition of the two electronic states. Our scheme is to launch a bound vibrational wavepacket in D_2^+ and, via manipulation of this wavepacket, to guide the molecule towards dissociation on both electronic surfaces, thus leading to electron localisation. This is done via a multi-pulse scheme, as sketched in Figure 2.

- First, a pump pulse (pulse 1) ionises the neutral deuterium molecule creating a vibrational wavepacket in the $1s\sigma_g$ ground electronic state of the deuterium molecular ion [6, 7, 8].
- A second time-delayed pulse (pulse 2) is applied to modify the vibrational wavepacket, redistributing the vibrational population [15, 16] and thus, inherently manipulating the wavepacket dynamics.
- The third pulse guides the modified wavepacket towards dissociation, localising the electron to one nucleus or the other.

The timing of both the second and third pulse interactions are set to coincide within a time window where the nuclear wavepacket is oscillating in a well localised manner i.e. during a vibrational revival.

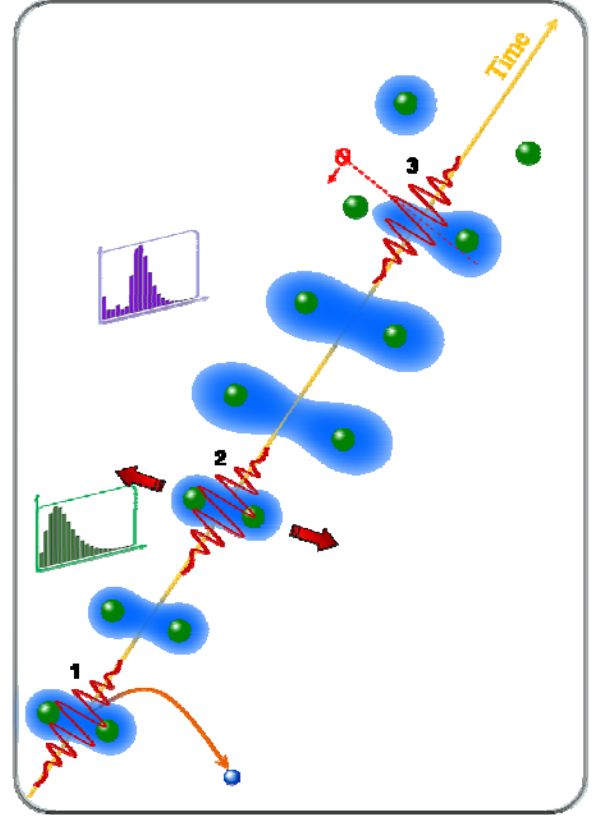


Figure 2. Multi-pulse scheme for enforcing electron localisation. Pulse 1 ionises the neutral D_2 molecule creating a vibrational distribution (green bar chart) in D_2^+ , pulse 2 is used to cause vibrational heating within the molecule (purple bar chart). This is subsequently fragmented via pulse 3, where tuning of its CEP controls which nuclei the electron localises to.

The control and dissociation dynamics are simulated using a quantum model which is well-established for vibrational wavepacket dynamics in D_2^+ [15]. The pump process is assumed to project the ground state wavefunction ($v=0$) of the neutral onto the ground electronic surface of the molecular ion. The resulting wavepacket is propagated within the time-dependent Schrodinger equation, via a Taylor expansion of the time-evolution operator. Subsequent pulse interactions with the vibrational wavepacket are incorporated in the Hamiltonian, expressed as a coupling between the two electronic states.

The characteristics of the control and dissociation pulses (7 fs FWHM duration, 2×10^{14} Wcm $^{-2}$ intensity) were chosen to reflect typical outputs from ultrashort pulse generation techniques, as well as to limit the onset of further ionisation [17], and were delayed with respect to the initial pump interaction over a range of delays in order to find the optimal conditions for electron localisation.

Results

Figure 3 displays results for optimised 2-pulse (b) and 3-pulse (d) simulations as a function of pulse CEP (ϕ), with the asymmetry summed over all dissociation events. There is a clear sinusoidal oscillation of the asymmetry as a function of ϕ . In the 2-pulse case a maximum asymmetry of 0.22 is achieved corresponding to a left: right ratio of just over 5:2. With the modified vibrational distribution fig 3(c) the asymmetry parameter reaches a maximum of 0.42 which equates to a left: right ratio of 4:1 and implies that the final position of the electron can be steered to the left in up to 80 % of the dissociation events during this pulse interaction.

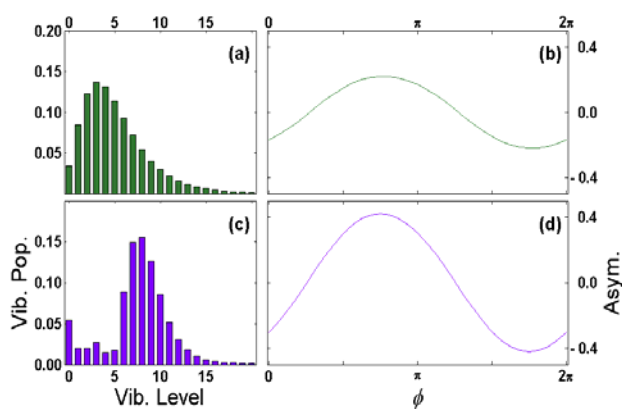


Figure 3. Asymmetry results for dissociation of a D_2^+ vibrational wavepacket. (a) Vibrational distribution created by the pump pulse. (b) Asymmetry as a function of CEP (ϕ) for dissociation of the distribution in (a), i.e. no control pulse applied. (c) Modified vibrational distribution created by a control pulse interaction at 598 fs. (d) Asymmetry for the modified vibrational wavepacket. For both (b) and (d) the timing of the dissociation pulse has been optimally chosen to enhance the dissociation probability and asymmetry.

In an experiment using apparatus such as a time of flight mass spectrometer [7, 8] or cold-target recoil-ion momentum spectroscopy device [9, 13] ions can be detected as a function of their kinetic energy release, constituting a key experimental observable. This can also be calculated from our results making it possible to observe how the asymmetry parameter varies as a function of the deuteron energy.

The kinetic energy release is shown in Figure 4 as a function of phase, for the optimised 3-pulse scenario where the colour scale denotes the strength of the asymmetry (red indicating the electron predominantly localising to the left nucleus and the blue indicating that it localises to the right). The figure indicates that controlled asymmetry is more likely to occur within certain energy ranges so while in this case the *total* asymmetry ratio is 4:1, within the energy range of 0.2 – 0.4 eV a peak ratio of 11:1 is observed meaning that 92% of the dissociation events that occur within this energy range can be manipulated by the pulse CEP. This is a significant enhancement on any previously observed asymmetry values.

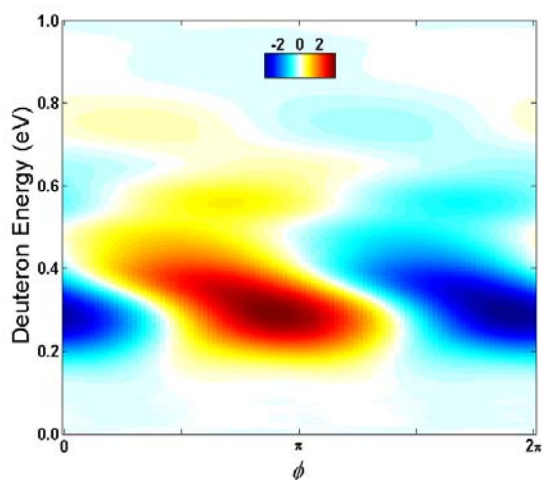


Figure 4. Electron asymmetry as a function of the dissociating pulse's CEP (ϕ) and kinetic energy of the dissociating deuterons. Colour scale denotes localisation where red = left nucleus and blue = right nucleus.

Conclusions

With current ultrashort laser technology, it is clear that we can turn our attention to the goal of controlling the motion of electrons in dissociating molecules. Before this is attempted in large molecules, it is useful to demonstrate the principle in simpler prototypal systems. Using established theoretical simulations we have identified an experimental configuration which uses the motion of a vibrational wavepacket and the pulse CEP to gate the electron localisation during photodissociation. By modifying the vibrational motion of the molecule, we can prepare the target into a suitable configuration for significant control to be exerted over the electron's final position. The localisation of the electron can be increased to 80% in all dissociation events and up to 92% within a 0.2 eV energy range.

Acknowledgements

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Intensity-resolved ionization processes in few-cycle strong-field laser pulses

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Introduction

In an intense laser field, ionization of atoms or molecules can proceed via the absorption of one or more photons, or by tunnelling due to distortion of the Coulomb potential by the incident electric field, as reviewed in^[1]. In this work, the authors concentrate on tunnel ionization only, as this is expected to be the dominant ionization process in few-cycle near-infrared (NIR) strong-field laser pulses. Keldysh^[2] was the first to attempt to quantify tunnelling, and his work has inspired several developments to the theory in the past fifty years^[3,4,5,6]. Most assume the outgoing electron can be considered separately to the parent ion after ionization, influenced only by the oscillating laser field. Corkum successfully explained non-sequential ionization processes such as recollision^[7] by treating the electron in this manner. Recollision occurs when in a linearly-polarized field the outgoing electron is accelerated back towards the parent ion within the same laser period which caused the initial ionization. The returning electron recombines with the parent ion or causes the removal of another electron. However, when these effects are suppressed, such as in the case of circularly-polarized pulses which send the outgoing electron on a trajectory which makes it unlikely to return to the parent ion, differences between theory and experiment are still seen. These differences have previously been attributed to excitation due to ‘shake-up’ by Bryan *et al.*^[8]. Due to the violent removal of the electron during an intense few-cycle pulse, higher-energy states with the same electron configuration can be populated. Alternatively, the population of higher-energy states could be explained by tunnel ionization directly to those states. It is also possible that lower-lying orbitals are involved in tunnelling, which have a much smaller but non-negligible ionization probability. Tunnel ionization from an orbital (HOMO-1) below the highest occupied molecular orbital (HOMO) has recently been experimentally observed in molecules by Akagi *et al.*^[9]. The ability to distinguish the orbital from which the electron has been ejected opens up the possibility of probing attosecond dynamics of individual electron orbitals.

A complete understanding of the tunnel ionization step is central to being able to control processes such as recollision, which is the first step in the production of attosecond extreme-ultraviolet (XUV) pulses by high-harmonic generation (HHG)^[7], tomographic reconstruction of electron orbitals^[10], and is used as the electronic ‘probe’ step in overlapped XUV-NIR experiments^[11,12]. There are also implications for molecular studies which use strong-field ultrashort pulses to initiate photodissociation and/or Coulomb explosion by ionization. An example would be the study and control of vibrational wavepackets, recently reviewed by Calvert *et al.*^[13].

We investigate tunnel ionization in xenon atoms using a few-cycle (~10 fs) NIR (700 – 900 nm), intense (10^{13} – 10^{15} W/cm²) laser pulse generated at Artemis using a hollow-fibre compressor^[14], which is temporally and spectrally characterized using frequency-resolved optical gating (FROG)^[15]. The pulse

is circularly polarized to suppress recollision effects. The ionization yield is measured with respect to distance from the focus, which will allow the yield to be intensity resolved. The results are compared to tunnelling models based on the theory of Yudin and Ivanov^[5], a popular theory in the attosecond community and is applicable to arbitrary values of the Keldysh parameter γ ^[2]. The proposed models take into account ionization between all possible combinations of inner- (5s) and outer-valence (5p) electron energy levels, and can be compared to ground-state only calculations to highlight the importance of their inclusion.

Eventually, the deconvolution method originally proposed by Van Woerkum *et al.*^[16,17] and further developed by Bryan *et al.*^[18] for a non-Gaussian focus will be used to extract ionization probabilities from the intensity-resolved experimental data, the results of which can be compared directly to calculated ionization probabilities. The deconvolution procedure relies on the correct characterization of the focal region. The laser focus is simulated by propagating an input beam profile through an ABCD system matrix. The input profile can be assumed to be Gaussian at the output of a typical laser resonator, but in the case of hollow-fibre compression is expected to be Bessel^[19]. Diffraction due to truncation would affect both beam profiles, but in the case of a truncated Bessel non-trivial focal distributions are anticipated^[20].

Modelling tunnel ionization

Bryan *et al.*^[8] demonstrated the importance of considering multiple electron energy levels in Ar tunnel ionization, finding better agreement between theory and experiment when multiple levels were included in the models used. The difference between multiple-energy-level and ground-state-only ionization was attributed to excitation caused by the outgoing electron, populating higher-energy ionic energy states. Recently, Akagi *et al.*^[9] identified ionization directly from a lower-lying molecular orbital (HOMO-1) in HCl due to tunnelling, with a lower but significant probability than the highest-occupied molecular orbital (HOMO). The HCl energy levels identified are of similar order to those in xenon, and their experiment uses similar laser intensities to the authors’ experiment.

Ionization probability strongly depends on the Keldysh parameter γ , which depends on the duration and intensity of the laser pulse, as well as the ionization potential I_p . For the removal of lower-lying electrons, I_p is much larger (in xenon, 23.4 eV for 5s removal, compared to 12.1 eV for ground-state 5p removal), and therefore the ionization probability is smaller, but not negligible. It may be possible for electrons to directly tunnel from lower-lying orbitals, leaving the ion in a higher-energy electron configuration. There are also multiple energy levels for each electron configuration (for Xe²⁺, there are five 5p⁻² levels in the range 33.1 – 37.6 eV^[21]) which are neglected when considering only ground-state ionization, tunnelling to each of which may contribute to the total ionization probability.

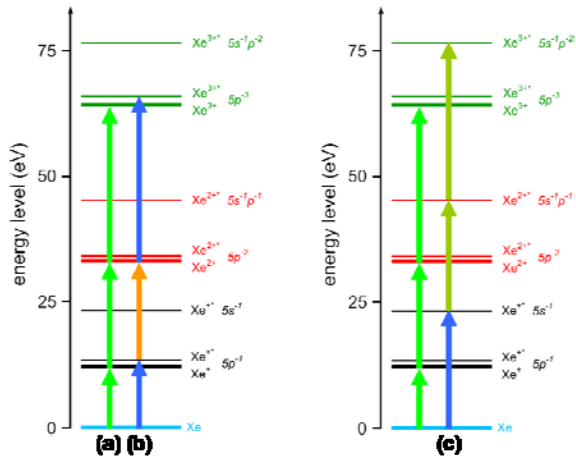


Figure 1: Valence energy levels in xenon, with example ionization pathways: (a) the outermost 5p electron is removed, creating ions in the ground state; (b) a lower-lying 5p electron is removed from the neutral atom to create an Xe^+ ion with a higher-energy electron configuration, which requires less energy to ionize further to Xe^{2+} ; (c) an inner-valence 5s electron is removed from the neutral atom with a much lower probability in Xe^+ , but ionization proceeds in subsequent charge states by 5p-electron removal, therefore contributing to the ionization

Figure 1 pictorially represents the effect of including multiple energy levels on the number and nature of the ionization pathways. Any differences in ionization probability with respect to ground-state ionization are expected to be small for Xe^+ , and increase with each subsequent charge state due to the increase in the number of available energy levels.

Ionization is modelled using the tunnelling rates calculated using the theory of Yudin and Ivanov^[5]. The resulting ionization probabilities are shown in Figure 2, which shows ground-state only and multiple 5p- and 5s-level ionization. Allowing tunnelling from the ground-state and next-lowest-lying 5p state introduces a small shift in the ionization probability. However, it can be seen that allowing for the cumulative effect of ionization from all 5p- and 5s-levels results in a substantial shift in ionization probability to lower intensities. The model takes into account ionization from all possible combinations of levels, hence the probability shifts become larger with increasing charge state.

Experimental setup

Figure 3 shows the layout of the experiment. A Femtolasers CompactPro Ti:S laser system produces 1 mJ, 30 fs NIR pulses which are spectrally broadened by self-phase modulation in an argon-filled hollow fibre^[19], and recompressed to ~10 fs by a series of chirped mirrors which compensate for the group-delay dispersion acquired after propagation through the fibre. A quarter-wave plate converts the pulse from linear to circular polarization. The pulse is reflectively focused ($f = 50$ mm) into an effusive xenon gas jet, generating intensities of $10^{13} - 10^{15} \text{ W/cm}^2$ over the focal region. The ions produced are detected using an ion time-of-flight (TOF) mass spectrometer, with a 250 μm aperture to limit the z -range, and therefore the range of intensities over which the ions are detected. The focusing mirror is varied with 100 μm resolution, translating the laser focus with respect to the aperture in the TOF. As a result, z -scans of ion yield vs. mirror position can be produced.

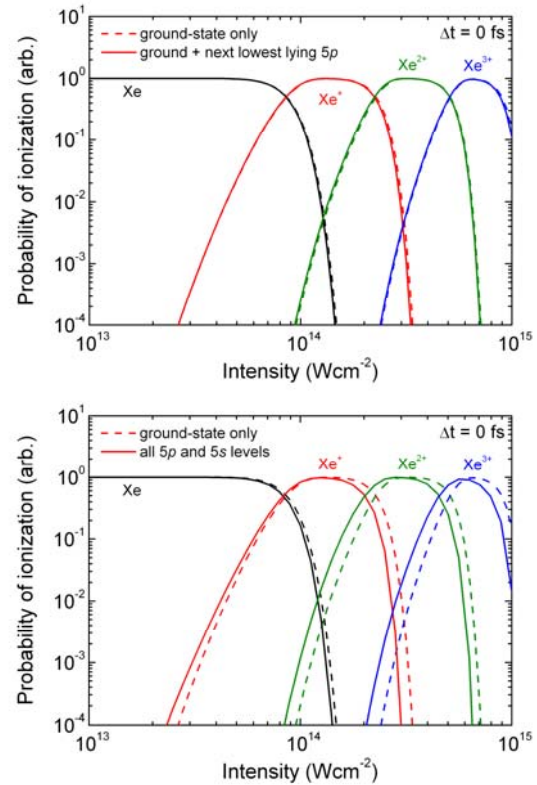


Figure 2: Predicted ionization probabilities from the theory of Yudin and Ivanov; (top) ground-state only ionization (dashed line) and ionization from the ground-state and next-lowest-lying 5p state only (solid line); (bottom) ground-state only ionization (dashed line) and ionization from all 5p and 5s states (solid line).

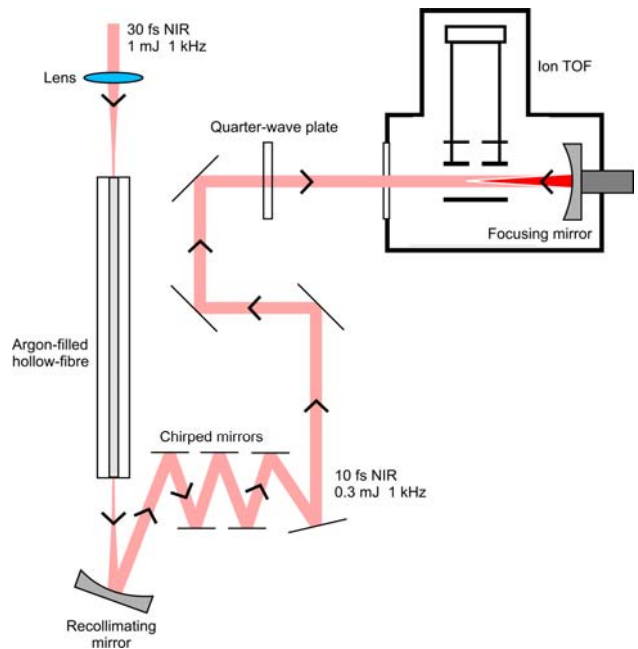


Figure 3: Experimental setup for temporally-shaped ultrashort NIR pulses. An argon-filled hollow fibre and chirped mirrors compress the initially 30 fs pulse to ~10 fs. The focusing mirror can be translated with respect to the TOF to spatially resolve ion production over the focal region. An effusive xenon gas jet (not shown) is situated near the focus.

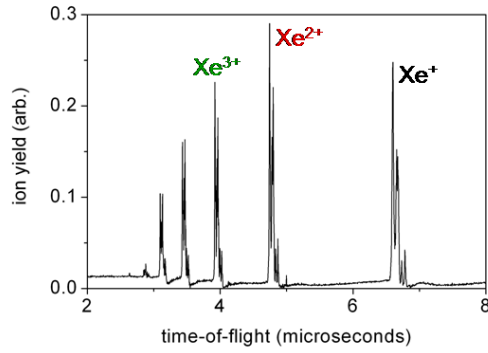


Figure 4: Xenon ion TOF spectrum, with the first three charge states identified.

Results

The xenon TOF spectra, an example of which can be seen in Figure 4, consist of ion yields for Xe^+ to Xe^{8+} separated due to their charge-to-mass ratio. TOF spectra are recorded for a range of distances from the focus, z . The ion yields for each charge state can be integrated, and plotted with respect to distance from the focus, producing z -scans. For successive charge states, the atomic ion requires higher intensities to ionize further, corresponding to a decrease in the signal-producing volume. This can be seen in the z -scans shown in Figure 5. Saturation behaviour is seen in the ion yields, most prominently in Xe^+ , where the yield depletes due to the probability of ionization in the next-highest charge state becoming significant.

The deconvolution^[18] procedure to extract ionization probabilities directly from z -scans is outlined in Figure 6. The on-axis intensity distribution is simulated over the same distance range as the z -scans by propagating an input beam through an ABCD optical system matrix representing the actual experimental setup in Figure 3. The deconvolution routine takes the on-axis intensity and integrated ion yield as inputs, and produces an ionization probability. The beam width and divergence at the input is not known precisely, and becomes the fitting parameter which is varied until good agreement is found between the theory and the deconvoluted ionization probabilities for the lowest charge state (Xe^+), which should be least affected by any excitation processes.

In order to verify the correct working of the deconvolution procedure, a three-dimensional ion-signal-producing volume has been simulated assuming a Gaussian intensity distribution at the input to the optical system matrix, with the parameters adjusted so that the features are on a similar scale to those of the experiment. Simulated z -scans were produced by integrating over a cylinder (250 μm radius) of the simulated volume over the same range of z used in the experiment, echoing the way in

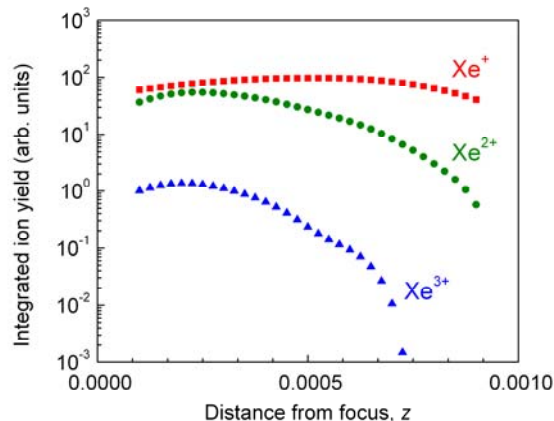


Figure 5: Experimental z -scans of integrated ion yield with respect to distance from the focus.

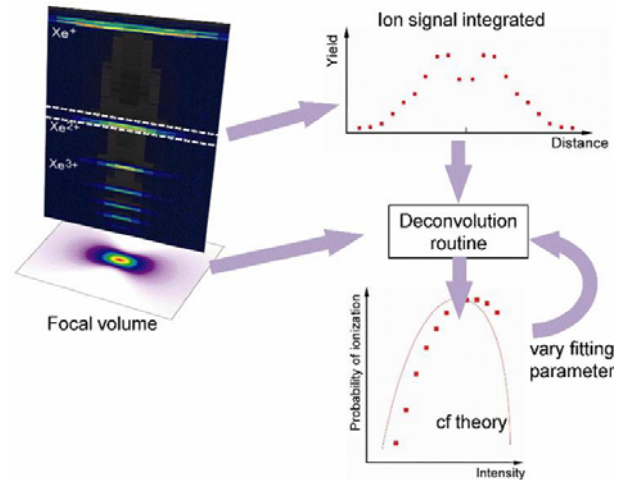


Figure 6: Deconvolution procedure to extract ionization probabilities from experimental z -scans.

which the ion yield is recorded by the apertured TOF. Since the simulated ion yields follow the predicted ionization probabilities for multiple electron levels as shown in Figure 2 (multiple $5p$ and $5s$ ionization), deconvoluting the simulated z -scans with respect to the simulated on-axis intensity distribution is expected to reproduce these probabilities, the results of which are shown in Figure 7. Apart from slight variations attributed to quantization of the signal volume in the simulation, the original input ionization probabilities are reproduced by the deconvolution.

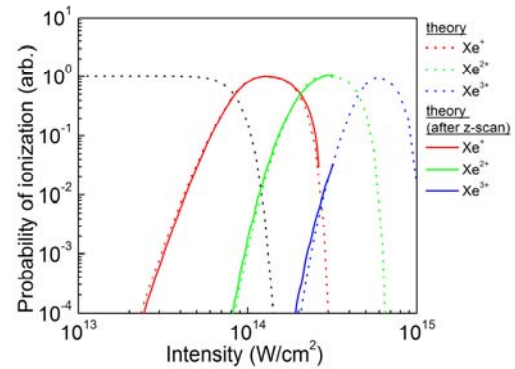


Figure 7: Predicted ionization probability before (dashed lines) and after (solid lines) being extracted from a simulated three-dimensional Gaussian ion-producing volume by deconvolution.

Conclusions

The recollision-free ionization of xenon in a NIR strong-field few-cycle laser pulse has been measured experimentally. Numerical models have been developed to take into account the contribution of multiple energy levels to the total tunnel ionization probability. The deconvolution procedure has been tested for a Gaussian input profile. Full characterization of the laser focus for a Bessel input profile is underway. The experimental ionization probabilities will be extracted from the data using the deconvolution method described earlier, the results of which will be the subject of a forthcoming publication.

Acknowledgements

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Ultrafast dynamics of electronic structure in complex materials

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Introduction

A fascinating variety of ordered phases emerge in systems with large, direct correlations between individual electrons [1], but their inherent complexity and the fact that diverse interactions take place on similar energy scales mean that a great deal of experimental input is often necessary. Invaluable techniques have included Angle-Resolved Photoemission Spectroscopy (ARPES), which maps out electronic band structure as a function of energy and momentum [2], and pump-probe studies, in which a transient excited state is produced by a femtosecond laser pulse and then studied by a second pulse after some delay [3]. Recently these techniques were combined to study the electronic structure of a transient phase using ultrashort UV pulses [4]. The new materials science beamline at Artemis has been commissioned to carry out this type of experiment. By employing high-harmonic pulses to generate photoelectrons, it offers the possibility of measuring time-resolved photoelectron spectra at momenta across the entire Brillouin zone. The test case for this new apparatus has been the layered sulphide 1T-TaS₂, which exhibits a range of phenomena of current interest.

1T-TaS₂

When cooled, this material undergoes a series of phase transitions as long-wavelength modulations in charge density form within the two-dimensional layers that make up its structure. At 180 K the Charge Density Wave (CDW) becomes commensurate and ‘locked in’ to the lattice, producing a static distortion to the charge density and atomic positions. The Ta ions form star-shaped clusters of 13 ions each. The resulting change in electronic structure removes 12 out of 13 electrons from the conduction band [5]. Cluster formation also increases the Coulomb interaction between remaining conduction-band electrons, causing localization and the opening of a Mott gap. Photoexcitation can destroy the Mott order and simultaneously excite an amplitude mode of the CDW. Optical measurements of the transient phase [6] reveal the combined effect of these processes. Time-resolved ARPES measurements at 6 eV showed a reduction of spectral intensity in the Lower Hubbard band (LHB) at the zone centre ($k=0$), corresponding to collapse of the Mott gap, followed by oscillation in the position of the band edge due to the CDW amplitude mode [4].

TR-ARPES on TaS₂ at Artemis

The measurements we show here are angle-integrated to include contributions from the entire folded Brillouin zone. Fig. 1 shows a map of photoelectron intensity as a function of energy and time. Immediately after photoexcitation with a 30 fs pulse of 1.5 eV light there is a collapse in intensity in the LHB, transfer of spectral weight towards the Fermi level, and subsequent oscillation of the LHB profile at the frequency of the CDW amplitude mode. These features agree well with those seen at $k=0$ [4], but our angle-integrated measurements additionally show an oscillation in the LHB peak intensity (Fig. 2), a new effect which we tentatively attribute to the influence of the CDW amplitude mode on the peak density of states for

momenta near $k=\pi/a$. In further experiments we will study the detailed dispersion of the dynamical response.

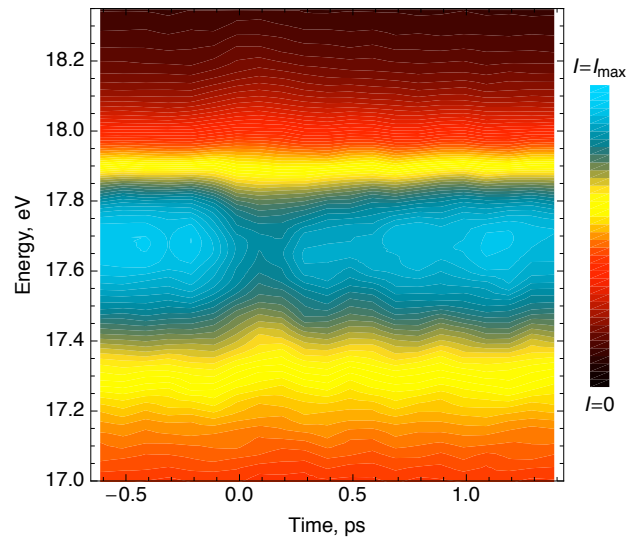


Fig. 1: Photoelectron intensity map of response to photoexcitation.

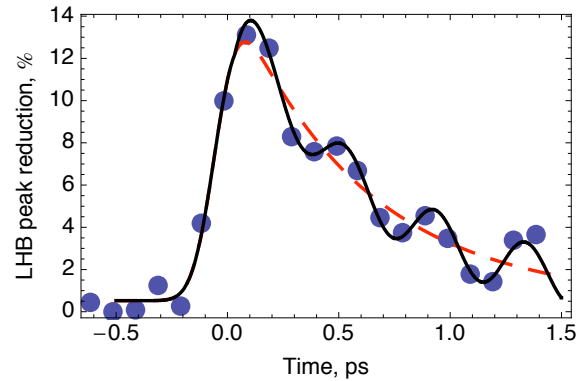


Fig. 2: Reduction in LHB intensity. Solid line: Full fit. Dashed line: Exponential component only.

Final Remarks

Artemis is uniquely suited to the task of understanding the relationship between CDW and Mott order in this material, and this will serve as an ideal first demonstration of the exciting possibilities offered by this new beam line.

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Ultrafast time-resolved photoelectron imaging of excited state molecular dynamics: S_2 - S_1 internal conversion in the DABCO molecule

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Introduction

The photodynamics of polyatomic molecules generally involves complex intramolecular processes which rapidly redistribute both charge and vibrational energy within the molecule. The coupling of vibrational and electronic degrees of freedom (resulting from the breakdown of the Born-Oppenheimer approximation) leads to radiationless processes such as internal conversion, isomerization, proton and electron transfer etc. These non-adiabatic dynamics underpin the photochemistry of almost all polyatomic molecules and are important in photobiological processes such as vision and photosynthesis, and underlie many concepts in active molecular electronics. If progress is to be made toward understanding and controlling molecular functionality it is clearly of fundamental importance to develop techniques to study and control non-adiabatic processes in complex molecular systems.

By virtue of its sensitivity to both electronic and nuclear configurations, femtosecond pump-probe time-resolved photoelectron spectroscopy has been shown to be a powerful and versatile probe of excited state photophysics. This technique allows electronic and nuclear motions to be disentangled, and in principle leaves no configuration of the molecule unobserved since ionization may occur for all molecular configurations [1]. This is in contrast to other techniques, such as absorption or fluorescence spectroscopy, which sample only certain areas of the potential energy surfaces involved, as dictated by oscillator strengths, selection rules and Franck-Condon factors. While a substantial amount of information is available from the evolution of the photoelectron energy spectrum in such experiments, significantly more information regarding the evolution of the molecular electronic symmetry is available from the photoelectron angular distribution (PAD) [1, 2] as measured in time-resolved photoelectron imaging (TRPEI) experiments. Here we report TRPEI measurements of the S_2 - S_1 internal conversion dynamics in the 1,4-diazabicyclo[2.2.2]octane (DABCO – see Figure 1) molecule following photoexcitation to the S_2 state. Studies at different excitation wavelengths have allowed us to examine the dependence of the electronic relaxation dynamics upon vibrational energy of the molecule.

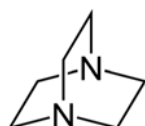


Figure 1: The DABCO molecule

Experimental setup

The experiments reported here were conducted at the Artemis facility at the Rutherford Appleton Laboratory. A schematic of the experiment is shown in Figure 2. TRPEI measurements were made using a velocity map imaging (VMI) photoelectron imaging spectrometer [3] equipped with a time and position sensitive delay line anode detector (Roentdek DLD40). An effusive inlet delivered DABCO vapour to the interaction region. During experiments the detector chamber pressure was maintained below 10^{-7} torr ensuring collisionless conditions.

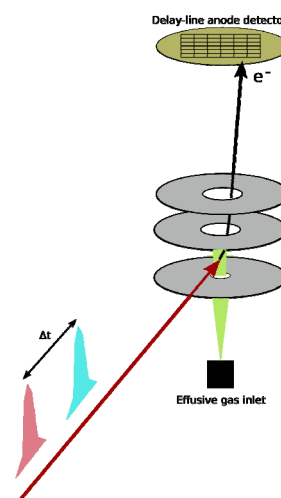


Figure 2: Schematic drawing of experiment. Pump and probe laser pulses interact with gaseous DABCO in the interaction point of a velocity map imaging spectrometer. The resulting electrons are accelerated to a time and position sensitive detector.

The pump and probe laser pulses were derived from a 1 kHz Ti:Sapphire CPA system (KM Labs Red Dragon) operating at 780 nm and delivering 30 fs pulses with 13 mJ pulse energy. The output of this laser system was split to produce synchronized pump and probe pulses. Tunable pump pulses in the region 236-252 nm were generated using a commercial OPA (Lightwave HE-TOPAS) and subsequent frequency mixing (signal plus pump followed by second harmonic generation) producing pulses with a duration of 40-50 fs and pulse energies up to 35 μ J. Probe pulses at 390 nm were obtained from frequency doubling of the output of the CPA system. For experiments employing a two-photon probe process, the output of the CPA system was used directly. Pump

and probe pulse energies were reduced to < 100 nJ using polarizers and half-wave plates as needed. Pump and probe pulses were recombined using an uncoated fused-silica flat with the UV pump pulse derived from the front surface (4%) reflection, and the probe pulse transmitted through the optic. Under these conditions on average less than one electron was produced per laser shot. The pump-probe time delay was adjusted using a motorized delay stage under computer control. Photoelectron images were obtained by collecting data for $0.25\text{--}2 \times 10^6$ laser shots.

Results

Figure 3 shows a representative photoelectron image obtained with a pump wavelength of 236.1 nm and single-photon ionization probe at 390 nm. Two distinct photoelectron bands are seen corresponding to ionization from the S_2 and S_1 electronic states of the molecule. Images were recorded as a function of pump-probe time delay, allowing us to follow the evolution from the S_2 state to the S_1 state. Figure 4 shows representative data of the population evolution in the two states obtained from a series of images at different time delays. In addition, we have information on the temporal evolution of the photoelectron angular distributions (not shown). Data was recorded for different pump wavelengths in the region 236.1–251.1 nm allowing us to examine the influence on the electronic dynamics of vibrational excitation in the S_2 state. In addition data was taken employing a two-photon probe ionization scheme at 780 nm. Analysis and interpretation of this data is ongoing.

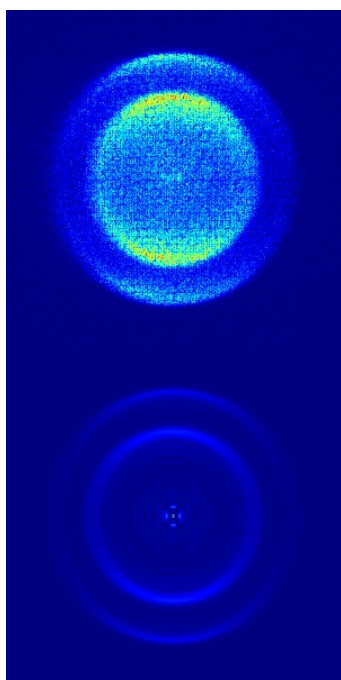


Figure 3: Photoelectron velocity map image recorded with a pump pulse at 236.1 nm and probe pulse at 390 nm. The raw experimental data is shown at the top, and the Abel inverted data is shown below. The outer ring corresponds to photoelectrons from ionization of the S_2 electronic state, and the inner ring corresponds to ionization of the S_1 electronic state.

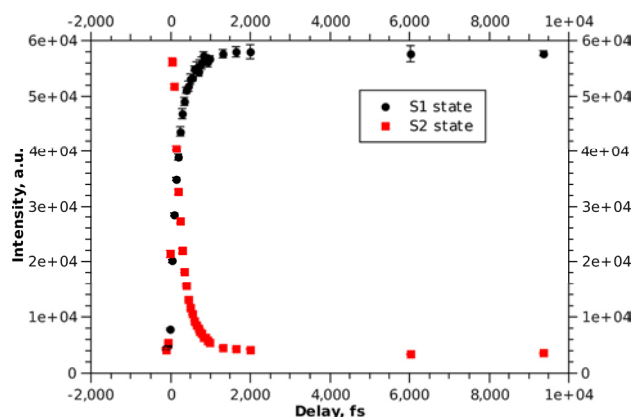


Figure 4: Populations of the S_2 and S_1 states as a function of pump-probe time delay following pump excitation at 236.1 nm.

Conclusions

Ultrafast time-resolved photoelectron imaging was applied to the study of internal conversion dynamics in the DABCO molecule as a function of internal energy. Analysis is ongoing.

Acknowledgements

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Probing metal complex dissociation in cells by fluorescence microscopy

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Introduction

SPECT and PET agents based on gamma or positron metallic radionuclides are in widespread use for the diagnosis of a number of diseases whilst beta or auger electron emitting radionuclides are used for therapy. Some important classes of imaging agent depend on intracellular processes to achieve selectivity, however the resolution limit for nuclear medicine of 1-2mm means that little is known about their speciation within living cells. Confocal fluorescence microscopy has been used extensively to track compounds and follow processes in cells. Subsequently we have designed a series of fluorescent metal complexes which can be used for whole body imaging using gamma or positron emission and followed in cells by virtue of their 1 or 2-photon fluorescence. We are particularly interested in the complexes of metallic radionuclides where a key step in their biological action can be dissociation of the complex within cells. The emission lifetime and emission wavelength of a ligand changes when a metal is coordinated and this provides a potential method to determine when and where dissociation of a metal complex occurs within a cell. We have explored the use of this approach to determine if and when intracellular demetallation of biologically active metal complexes occurs. The chemical series of current interest, design and research within this collaboration is; copper bis(thiosemicarbazone) complexes, indium tridentate thiosemicarbazones and indium porphyrins.

Copper bis(thiosemicarbazone) complexes

Bis(thiosemicarbazone) complexes of 64-copper(II) such as CuATSM (Figure 1) are in current clinical use for the imaging of hypoxia¹ although their performance is far from ideal. The hypoxia selectivity depends on the reduction of the complexes in hypoxic cells followed by dissociation and trapping of the copper. However, very little is known about the dissociative process and as the simple CuATSM complex is not fluorescent we have attached potent fluorophores (i.e. bodipy, fluorescein and anthracene) to the CuATSM core in order to follow the fate of the complexes in cells, Figure 2.

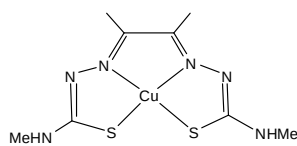


Figure 1: CuATSM

The fluorescence emission data for the anthracene conjugates show that the free ligand and Cu complex emit at very distinct wavelengths, 510 nm and 450 nm respectively, which means that it should be possible to distinguish them in cells by the spectral analysis of the emitted fluorescence. The emission lifetime data for the bodipy and fluorescein conjugates suggest that it is, in principle, possible to discriminate between free ligand and copper complexes in cells. In the case of the bodipy adducts a change in lifetime from 4.1 ns for the free ligand to a lifetime of 2.9 ns for the copper complex is observed, which

allows a level of discrimination between the two species *in vitro*.

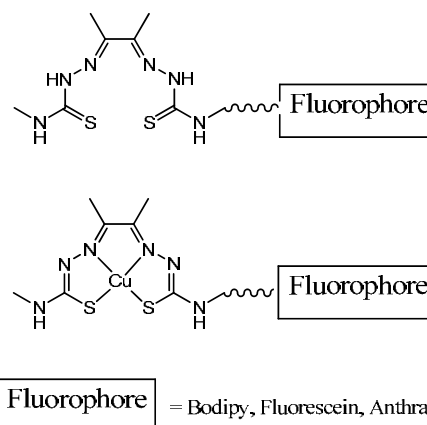


Figure 2: Structures of fluorescent CuATSM adducts

Figure 3 shows lifetime emission maps for the free bodipy/ATSM ligand and the Cu complex in HeLa cells after 1 h incubation with the data suggesting that the Cu complex does not dissociate over time. This experiment was conducted under normoxic conditions so the lack of dissociation *via* reduction is not unexpected. We are currently investigating the hypoxia selectivity of the conjugate *via* 64-copper radiolabelling and will conduct the cell experiments under hypoxic conditions to establish quantification of the degree of dissociation.

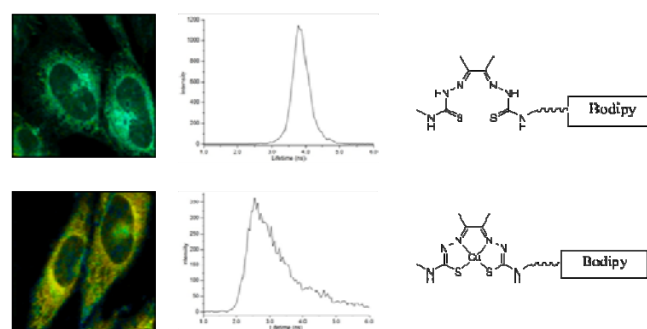


Figure 3: Emission lifetime maps for free bodipy/ATSM ligand(1) (top) and copper complex(3) (bottom).

Furthermore, new Group 11, 12 and 13 metallic bis(thiosemicarbazonato) complexes with extremely highly kinetic stabilities in serum have also been investigated as potential alternative synthetic platforms for the simultaneous diagnosis and therapy of cancer.^{2,3} This new family of intrinsically fluorescent metal bis(thiosemicarbazones) is shown in Figure 4 below, where M = Zn(II), Cu (II), Ga(III)Cl, In(III) and R = Me, Et, Ph and Allyl. Two photon spectroscopy (λ_{ex} 910 nm) was recorded in a variety of solvents and concentrations on the ligands and corresponding complexes. These complexes showed potent cytotoxicity in a number of

cell lines, coupled with interesting coordination modes for the ligand i.e. a symmetric and asymmetric mode, as shown by X-ray diffraction for $M=\text{Zn(II)}$.²

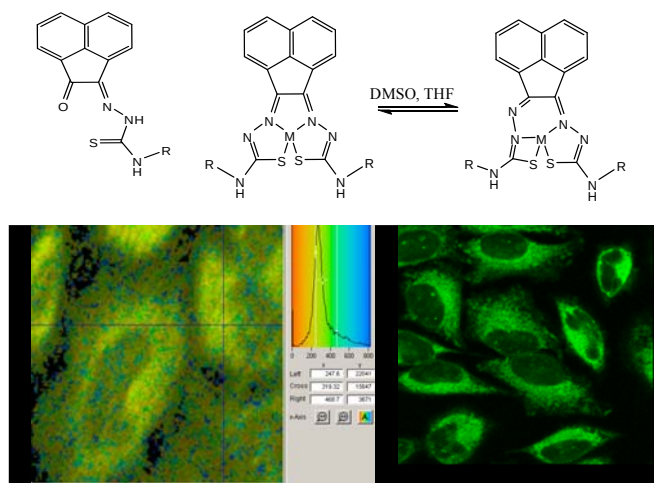


Figure 4: Representations of the aromatic bis(thiosemicarbazones) studied, and preliminary proof-of-concept FLIM (left) and confocal imaging in HeLa (right) for a Zn(II) analogue

The symmetric and asymmetric complexes are in exchange in solution as evidenced by ^1H NMR techniques and we are now hoping to use fluorescence spectroscopy to identify the nature of the compound when taken up by the cells and whether or not decomposition to free ligand would be observed. The closely related free ligand shown above is only very weakly fluorescent with ca 478 ps lifetime in solution and ca 497 ps lifetime inside the HeLa cell cytoplasm. The confocal fluorescence (λ_{ex} 488/ λ_{em} 543 nm) of the corresponding Zn(II) complex investigated in DMSO showed ca 302 ps lifetime (TCSPC in solution, 50 μM) and strong uptake in HeLa cells with ca 307 ps lifetime inside the cytoplasm (60 mins uptake). We are now studying derivatives with a variety of substituents at the exocyclic nitrogens and exploring the possibility that the Group 13 derivatives localise in the cell nucleus. Our preliminary data suggested that the Zn(II) complex is highly kinetically stable in cells and that this emerging spectroscopic technique may allow us to observe dissociation /decomposition of complexes in cells via emission lifetimes.

Future work: Nuclear targeting complexes for therapy

There is currently much interest in the use of complexes of metallic radionuclides for therapy. A potentially attractive route is to use very short path length Auger electron emission to cause DNA damage however this requires localization of the metal within the nucleus to maximize dose efficiency. This approach has recently been used very successfully for a ^{111}In complex conjugated to h-EGF protein to target breast cancer and is currently in clinical trials.⁴ Currently confirmation of nuclear localization requires lysing and cellular fractionation.

Metal complexes of tridentate thiosemicarbazones

The thiosemicarbazone shown in Figure 5 is in clinical use under the name of Triapine as a broad spectrum anti-tumour agent and its activity is enhanced by coordination to metal ions as an NNS donor.⁵ 64-Copper complexes of related thiosemicarbazones have been shown to have high uptake in tumours and have potential as tumour imaging agents.⁶ Despite their clinical use there is little known about the speciation of such metal complexes in cells and we have developed some fluorescent analogues to probe their behaviour in cells.⁷ These complexes are potent ribonuclease reductase inhibitors⁸ and would therefore be expected to localise in the nucleus. The structure of the In complex of a fluorescent quinolyl analogue L

which has retained potent cytotoxic behavior is shown in Figure 5

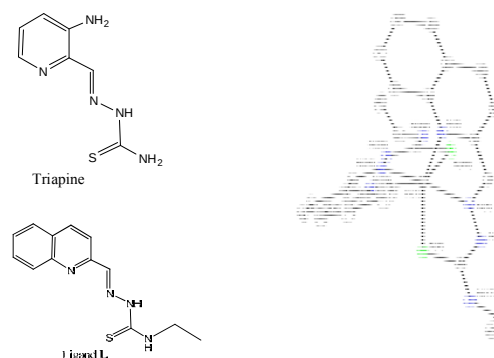


Figure 5: Structure of Quinolyl ligand L, Triapine and a fluorescent indium quinolyl complex InL_2Cl

Preliminary data on these species gathered at RAL suggests that it will be possible to observe dissociation in cells by means of both the wavelength of emission or emission lifetimes. These experiments will be conducted in the next testing sessions at the RAL.

Targeted Metallo-porphyrins

We have recently found that intrinsically fluorescent positively charged indium porphyrin complexes display nuclear uptake without addition of any nuclear targeting agent. Fluorescent images confirm nuclear uptake but do not discriminate between indium complex and free porphyrin. We will now use the change in emission lifetime and differences in emission wavelength between these to confirm if the indium metal is located in the nucleus. Comparison with other porphyrins suggests that the positive charges play an important role in directing the complexes to the nucleus.

Conclusions

Lifetime resolved 2-photon microscopy has clear potential as a tool for investigating the speciation of metal complexes in cells and *in viscera*. Much remains to be done, and we are currently focusing on probing the speciation of these complexes further, and on the development of new methods for image analysis and processing.

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Introduction

To engineer a complex 3D tissue from cells the production and utilisation of the appropriate 3D scaffold is critical. Cells presented with a flat surface grow typically in a monolayer fashion, while 3D cell cultures can only be achieved via growing them in a 3D micro-environment that is optimally tuned to enhance their adhesion and growth.¹

Thus the central research question in scaffold manufacture for tissue engineering is; *what specific microstructure and physical properties does a tissue scaffold need to present for optimal cell adhesion/growth?* The main approach is to try to reproduce the microstructure of the Extracellular Matrix (ECM) of a given tissue type. Indeed, the ECM, or the connective tissue between the cells, provides the necessary nanoscale chemical, structural and mechanical environment for a set of cells in a tissue.² Although we are still far away from exactly mimicking the ECM by a cell scaffold, important advances have been made in tissue engineering to provide an increasingly detailed substitute for the ECM.

3D tissue scaffolds have been produced as random or ordered microstructured networks. Random 3D networks are produced via e.g. solvent casting with particulate leaching, electrospinning or phase separation in combination with freeze-drying/critical point drying³ while free-form fabrication techniques (such as microstereolithography, μ SL) are used to make ordered 3D networks. In general random networks, when compared to ordered structures, are easier to produce in bulk but their microstructure and physical properties are more difficult to control, analyse and interpret. Studies based on scaffolds formed from random 3D networks dominate the current research due to their ease of manufacture when compared to the fabrication of ordered networks. Nevertheless, some recent high-profile studies have indicated a distinct advantage in user-controlled manufacture of 3D structures for tissue engineering. For example, Langer et al. have been able to grow directionally aligned heart muscle tissue for the first time, mimicking natural heart tissue, via using a tissue scaffold with an ordered microstructure.⁴ This study showed that the alignment of the heart muscle cells was critically dependent on the specific microstructure of the scaffold.

As indicated by this example; a very important advantage of controlled manufacture of 3D structure via free-form fabrication methods is that a controlled topological environment for cell growth can be provided. Since the 3D structure can be easily adjusted (via changing the structural parameters in a computer aided design (CAD) program), this manufacture platform is ideally suited for exploring the relationship between 3D topology and cell behaviour and to tease out the exact design criteria for scaffolds for different cell and tissue types.

Stereolithography (SL) enables fast prototyping of 3D objects through photopolymerisation. In the photocuring process, a mixture of photoinitiator and reactive prepolymers (monomers or oligomers) is exposed to intense light. The photons are absorbed by the photoinitiator which breaks down and initiates the reaction, forming an insoluble polymer from the prepolymers. The photoinitiators used in this experiment break down under UV light, thus in the standard SL set-up UV light is used to polymerise the material via a one-photon process. In one-photon polymerisation (1PP), the polymerisation typically takes place on the surface of the irradiated polymer and the

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polymerisation depth is governed by the Beers-Lambert law, which limits the in depth resolution. In the writing plane the resolution of this experiment is wavelength-limited and is seldom better than 1 μ m.

A novel route to photostructuring is via exploiting 2-photon polymerisation (2PP), where photons of double the wavelength of 1PP are used (typically $\sim$ 800 nm), so that the photoinitiator needs to absorb 2-photons to break down and initiate the polymerisation. Given that in 2PP absorption cross-section is much smaller much higher intensity light sources need to be used, and typically femtosecond lasers are used to achieve this. This revolutionary production process exhibits important advantages compared to methods based on 1PP, and allows the production of user defined (sub)micrometer definition structures. Indeed, the smallest feature size that has been reported via 800nm 2PP is $\sim$ 100 nm.⁵

In a former pilot study we have shown that with this set-up we can build 3D structures of 3-4 μ m resolution polycaprolactone polymer scaffolds with 2PP.⁶ In this study we report a further exploration of 2PP processing of polylactide based polymers.

Both of these polymers are biocompatible, biodegradable and tissue engineering scaffolds produced with these materials provide initial rigidity to an engineered tissue while the tissue builds its own scaffold, i.e. the Extracellular Matrix (ECM), and afterwards the engineered scaffold is resorbed by the tissue.

Experimental

In this project we have used a scanning μ SL set-up in which focused light is scanned through a liquid photocurable material via a high accuracy xyz-translation stage. The regions which are expose to the light cure or harden, unexposed region remains liquid and can be washed away, resulting in a 3D pattern.

In this project a laser direct write system was constructed which combined a Ti: sapphire laser (Coherent MIRA-900, 810nm, repetition rate 76MHz, pulse width 200fs) focused through a 20x 0.75NA lens (Nikon) with a high precision xyz-translation stage (Aerotech). For the 2 photon-polymerisation an *in house* synthesized photocurable PLA prepolymer (synthesis route described in Ref. 7) was used and this prepolymer 4,4'-bis(diethylamino)benzophenone (Sigma-Aldrich) was used as the photoinitiator for two-photon polymerization. The photoinitiator was used at 2 wt % to the pre-polymer.

Structures were created in an automated fashion by creating a program consisting of a series of linear translation instructions (e.g. x+3mm, y+50 μ m) for the translation stage. Samples were prepared with the resin sandwiched between a glass microscope slide and a 13mm circular glass coverslip separated by two bands of adhesive tape, and the structures written onto the face of the glass coverslip. The glass coverslips were treated with 40mM 3-methacryloxypropyltrimethoxysilane (MAPTMS) in dichloromethane (DCM) prior to use to aid structure adhesion. Indeed, when bare glass coverslips were used the produced microstructures did not adhere to the glass surface and floated off the glass surface during development. When fabrication was complete the samples were developed in toluene to remove uncured monomer and left to dry in air.

The produced structures were used to study the cell growth and attachment of NG108-15 cells, a hybridoma cell line of a rat neuroblastoma and a mouse glioma. This provides us an initial assessment of the neurocompatibility of these scaffolds. The produced structures were briefly sterilized by immersion in IMS

followed by soaking in PBS. NG108-15 Cells were seeded over a range of concentrations and cultured for 48h before being stained for fluorescence imaging.

Results

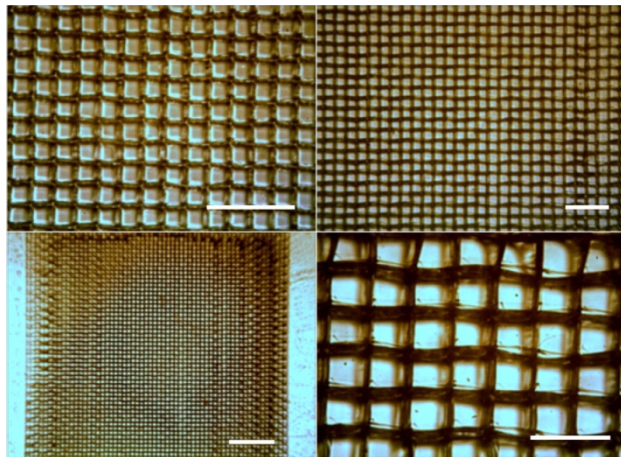


Figure 1: Optical micrographs of PLA microstructures created by laser direct write. Fabricated at a power of 1W and write speed of 5 mm s^{-1} . Scale bars: Top row 200 μm , bottom left 500 μm , bottom right 100 μm .

Our set of experiments explored the rapid fabrication of macroscopic 3D objects with 2PP. To ensure the polymerization was a true 2PP, we tested polymerizing the prepolymer-photoinitiator mixture under 1W, 810 nm, continuous wave light and no polymerization was observed. Only when the laser was mode-locked properly to provide femtosecond radiation polymerization was observed.

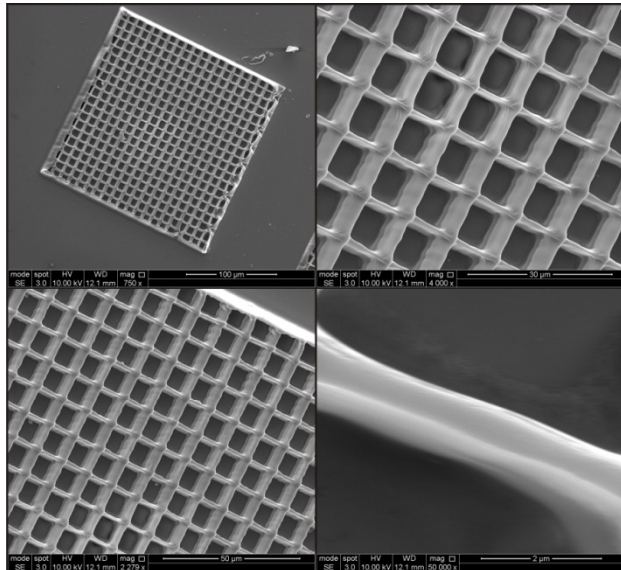


Figure 2: By expansion of beam filling objective feature resolution was reduced to less than $1 \mu\text{m}$. Scale bars: Top left 100 μm , top right 30 μm , bottom left 50 μm , bottom right 2 μm .

A series of experiments was carried out to determine the optimal write speed while fully exploiting the high beam intensity of the laser. Fig. 1 shows the optical microscopy photographs taken of the resulting structures. These structures were written with a beam intensity of 1W and with write speeds of 5 mm s^{-1} , and as shown in Fig. 1 considerably large structures can be achieved in a feasible timescale (5 mins). The line spacing of these cross-hatch structures is 50 μm and the thickness of the individual lines is $\sim 10 \mu\text{m}$. An important note is that some of the patterns look distinctly distorted (can be clearly

seen in the bottom left in Fig 1). This was mainly attributed to mechanical instabilities during the direct write process and this was largely improved upon during the project.

The thickness of the lines produced is dependent on write speed, beam power and also the cross sectional area of the focal point of the objective. When the back aperture of the objective is filled by the Gaussian-shape laser beam a more defined focal point results giving finer feature resolution. By using a telescopic beam expander (consisting of a 5 cm and a 15 cm lens) to expand the beam a resolution of less than $1 \mu\text{m}$ was achieved in the xy-direction. The obtained structures are given in Fig. 2, and in this case consist of a 200 μm cross-hatched pattern with line spacings of 10 μm . The resolution in the z-direction is 2–5 μm .

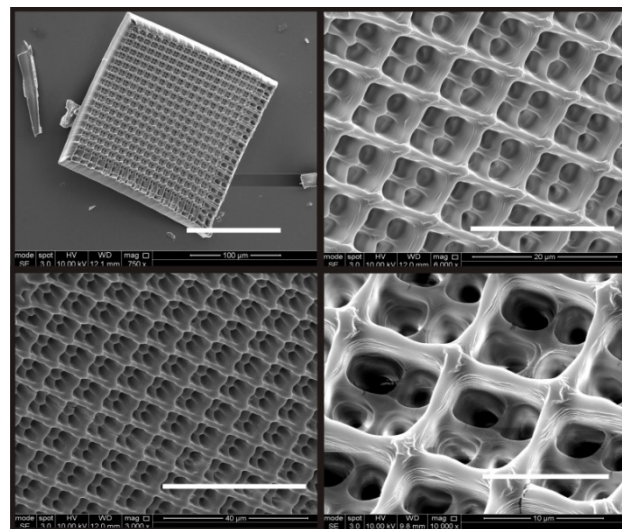


Figure 3: By writing more than one layer and offsetting the pattern three dimensional porous structures can be produced. Scale bars: Top left 100 μm , top right 20 μm , bottom left 40 μm , bottom right 10 μm .

An additional feature of 2PP, is that precise 3D microstructuring can be achieved. Indeed, the 2PP technique is up to date the only direct write technique that can deliver ‘real’ 3D micro- and nanostructuring. During this project we have investigated the possibility of 3D structuring via 2PP, and SEM micrographs of the obtained structures are presented in Fig. 3. In this set of experiments 2 cross-hatched patterns were constructed with a vertical (z-axis) offset by 2 μm and a xy offset of 5 μm (or half the line spacing). One can clearly observe from Fig. 3 that a 3D microporous structure was achieved with this set-up.

These structures, produced by 2PP, have also been studied for neuronal tissue engineering applications. In a first step we assessed the biocompatibility of this particular blend of PLA via studying the viability of human dermal fibroblasts on spin-coated thin films of this material (data not presented). From this data we could conclude that this material proved to be, in general, biocompatible. An interesting question that could be posed now is if the microstructuring can influence cell growth on this material. In particular, our group is interested in using this material for neural tissue engineering. To assess this we cultured neuronal type (NG108-15) cells cultured on 3 sets of produced PLA patterns in a pilot study. The structures consisted of $\sim 10 \mu\text{m}$ thick lines arranged into 3 mm square grids as detailed before. The patterns produced consisted of an array of squares with lines spaced by 50 μm centre to centre in x and y, an array of rectangles with 50 μm spacing in x as before and a spacing of 150 μm centre to centre in y, and an array of parallel lines with a centre to centre spacing of 50 μm . These patterns were chosen to investigate the effect of microenvironment on

cell growth of these neuronal cells. NG108-15 cells readily align and grow along linear micropattern,⁸ while this might not be the case on a square pattern. Indeed, when these cells were seeded on the scaffold and their attachment/growth was investigated after two days in vitro, one can observe that the cells only attached to the parallel lines, and failed to attach to the rectangular and square structures, indicating a degree of shape selectivity in their attachment and growth (see figure 4).

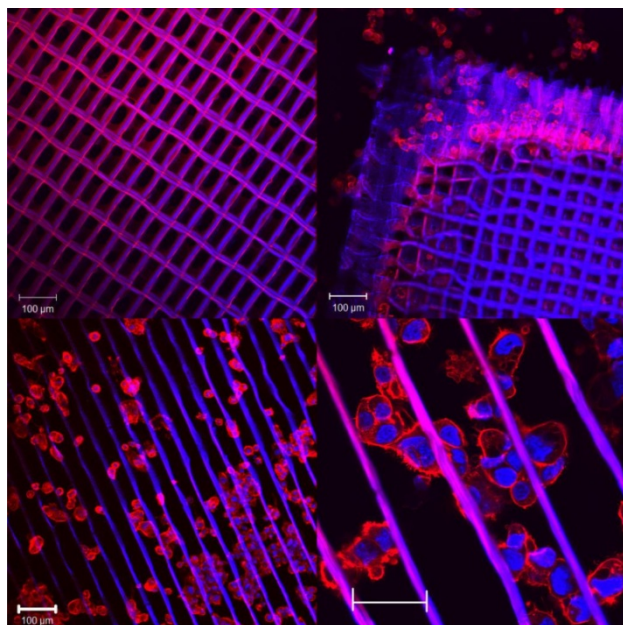


Figure 4: NG108-15 Cells on Microstructured Surfaces after 48h in culture. Cells have attached successfully to parallel lines of Microstructured PLA however did not attach to square or rectangular structures indicating a degree of surface selectivity. Scale bars top and left, 100 μ m, bottom right 10 μ m.

Conclusions

Scanning microstereolithography has been used to create a range of structures with feature resolution as low as 1 μ m from a photocurable, biocompatible and biodegradable polylactide formulation. The ability to create porous resorbable scaffolds with user defined microstructure was demonstrated, showing the potential of this technology for fabricating structures which mimic the extracellular matrix. Furthermore, it was demonstrated that microstructure can be used to influence cellular behavior, in this case attachment and alignment.

Acknowledgements

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Introduction

The Plant Endomembrane Group at Oxford Brookes has a long history in the study of the structure and function of the higher plant secretory pathway. This is the series of organelles in the cell responsible for the synthesis, processing and packaging of proteins, carbohydrates and lipids that are either sent to the vacuolar system for storage (or other functions) or to the cell surface for exocytosis. Over recent years the group has specialised on *in vivo* imaging and confocal microscopy of the secretory pathway using fluorescent protein constructs. We were the first to describe the motility of leaf Golgi bodies over the cortical endoplasmic reticulum (ER) network (Boevink *et al.* 1998). This initial success resulted in the award of a number of BBSRC grants looking at protein transport between the ER and Golgi and the nature of the ER export sites. Photobleaching technology has been used to characterise the transport of membrane proteins between the two organelles and recently the novel concept of the Golgi apparatus and the ER export sites working in tandem as unified complexes moving over or in the ER membrane has been published (Brandizzi *et al.* 2002, daSilva *et al.* 2004, Runions *et al.* 2006). This raises the intriguing question as to how individual Golgi bodies manage to stay tethered to the ER and yet to maintain the integrity of the stack whilst being propelled along the ER.

The successful functioning of the plant secretory pathway is also dependent on the sequential interactions of a myriad of proteins each with a defined task. Unraveling the interactions between various sub-sets of these proteins is a main goal of our research programme, which also involves a number of national and international collaborations.

This report summarises a series of experiments that were carried out to investigate the interactions between plant Golgi matrix proteins and regulatory GTPases (Latinjhouwers *et al.* 2005) as a continuation of the work reported in the CLF annual report 2006-2007. We have also carried out a series of experiments to investigate potential hetero- and homo-dimerisation of plant reticulon proteins that are responsible for maintaining the curvature of endoplasmic reticulum tubules.

Protein Interactions at the plant Golgi apparatus

Golgins are large coiled-coil domain proteins and in animal cells it has been shown that they can dimerise or interact with other golgins to carry out tethering functions within the secretory pathway. Therefore GFP fusions of AtCASP or GC2 (a homologue of the animal golgin-84 predicted to be located at the *cis*-Golgi) respectively were expressed on their own to determine their lifetime and the obtained GFP lifetimes were compared to the ones of GFP co-expressed with mRFP fusions of the same protein. No significant reductions in GFP lifetimes were observed for the combinations GFP-AtCASP with mRFP-CASP and GFP-GC2 with mRFP-GC2. In a previous study we had determined that GFP-AtCASP and mRFP-GC1 did not appear to interact *in vivo*. Therefore we now tested GFP-AtCASP with mRFP-GC2 but did not observe any interaction either. This indicates that the functions and tethering mechanisms of plant golgin homologues might work in a different manner compared to their counterparts in animal cells.

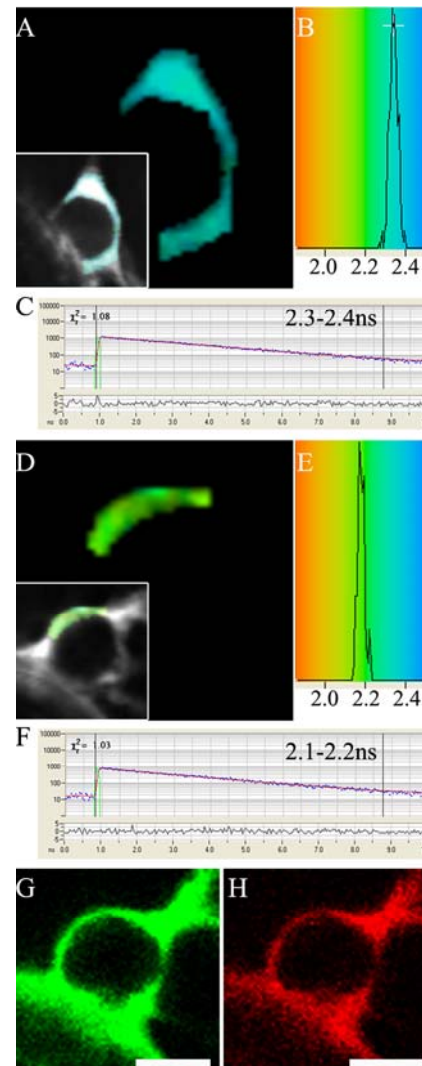


Figure 1. FRET-FLIM analysis of RTNLB1 dimerisation in tobacco leaf epidermal cells.

RTNLB1-eGFP (A-C) and coexpression with mRFP-RTNLB1 (D-H) in tobacco leaf epidermal cells were tested for interaction through FRET-FLIM. A region of interest on the ER continuous with the nuclear envelope was selected (A,D, see inset) based on the lifetime decay curves of points within the region having a χ^2 value between 0.9-1.4 (C,F). The lifetimes of the points within the selected region are displayed (B,E) and pseudocoloured accordingly in A and D. Confirmation of coexpression of eGFP and mRFP RTNLB1 fusions are shown (G,H). The lifetime of the combination is more than 0.1ns lower than the donor fusion alone, indicating RTNLB1 dimerisation. Scale bar 5 μ m.

Many mammalian golgins also bind to small regulatory GTPases. These interactions are thought to be required for the correct Golgi stack localisation of golgins as well as for trafficking and membrane tethering events. We have previously proved this to be true for some *trans*-Golgi golgins (Hawes & Osterrieder 2007, Osterrieder et al. 2009). We now tested GFP fusions of the *cis*-Golgi located Golgins AtCASP, GC1 and GC2 with mRFP fusions to the small plant GTPases RabD2A, RabB1b, Rab-H1^b and Rab-H1^c. Surprisingly, no significant decrease of GFP lifetimes was observed for any of the combinations, which suggests that these proteins might employ different Golgi targeting mechanisms or might interact with other small GTPases than the ones studied in this screen. Further work is being undertaken with other *cis*-Golgi golgins such as p115.

Protein interactions in at the plant endoplasmic reticulum

The endoplasmic reticulum (ER) is the key organelle for producing and processing proteins for storage or export from the cell. As part of an on going project to investigate the organisation of the cortical ER network we have been studying the role of the reticulon family of proteins in inducing curvature of the ER network (Sparkes et al. 2010). We have demonstrated that these proteins are restricted in distribution to membrane tubules and curved edges of cisternae and not to the faces of cisternal sheets. One hypothesis is that they form a "W" shaped wedge in the ER membrane and either hetero- or homodimerise or oligomerise to induce curvature. Interactions of various combinations of reticulons and truncated forms were measured by FRET/FLIM in cortical epidermal cells of tobacco leaves, after transient expression of green and red-fluorescent protein tagged constructs (Sparkes et al. 2010). The average of the range of pairs of constructs expressed indicated at least a 0.2 ns decrease in lifetime of the pairs compared to the control donor expressed alone. Therefore, all combinations of full-length RTN and truncations tested appeared to show some degree of oligomerisation.

Development of Laser Tweezers on a TIRF microscope

Our initial attempts at TIRF microscopy gave totally unexpected results (Hawes et al. 2010). In theory the presence of the cell wall in plant tissue precludes the use of TIRF as a

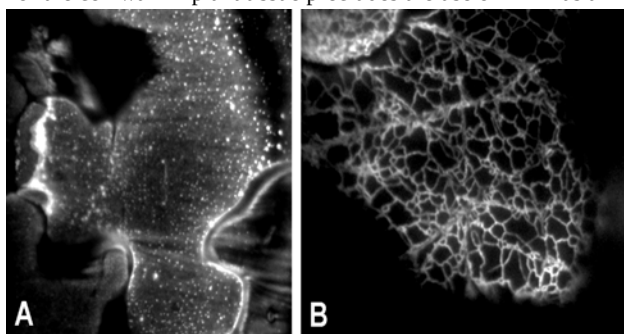


Figure 2. A. TIRF image of the plasma membrane of a tobacco leaf epidermal cell expressing GFP-LTI6B showing punctae and a filament associated with the PM (image courtesy of A. Martiniere). B. TIRF image showing cortical endoplasmic reticulum in a similar cells labelled with GFP-HDEL.

viable technique. However, in our first trial runs stunning images of protein location on the plasma membrane, of cortical ER and Golgi bodies were obtained. We assume that this is due to the cell wall having a similar refractive index to coverslip glass and thus the evanescent wave occurs at the wall/plasma membrane interface (Hawes et al. 2010). This has opened up a whole new range of experiments on secretion to the plasma membrane. As a result of these data it was decided to build laser tweezers into a TIRF microscope rather than the confocal attached to the two photon FLIM set up. This is based around a Nikon TE-200-S inverted TIRF microscope with a 1.49NA oil

immersion objective. Imaging is carried out with a 473nm diode laser and trapping with a 1064 Nd:YAG diode laser focussed to the imaging plane of the microscope. This development work has been carried out by Dr Andy Ward and a sandwich student Alasdair McKenzie and the first successful trapping of fluorescent beads has just been carried out prior to experiments being undertaken on the trapping of Golgi bodies in tobacco and arabidopsis leaves.

Conclusions

Work on the interactions of Golgi matrix proteins with small regulatory GTPases and with each other is on going, further combinations of proteins are to be tested. FRET/FLIM can be used to study self interactions of proteins on the endoplasmic reticulum and this work will be extended to cover interactions of ER-plasma membrane anchor point proteins as they are identified in other projects. TIRF is a technique that can be used with plant tissues and as such opens up a new avenue for studying interactions at the plasma membrane. Also the major components of the cortical endomembrane system can be imaged which will permit the use of a combination of laser trapping and TIRF microscopy to manipulate organelles such as the Golgi apparatus.

Acknowledgements

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Supra-molecular rules in signalling networks: A single molecule comparative study in cells and tissues

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Introduction

We have developed single molecule and other fluorescence techniques on the Octopus facility, combined with simulation, modelling and analysis techniques as part of a programme in the Lasers for Science Facility (LSF). Our first aim is to define patterns of conformation association, activation and signal transduction for the 4 members of the ErbB family and their correlation with plasma membrane proximal receptor traffic and signalling using cultured cells. We also aim at deriving models of ligand-induced behaviour using single molecule and ensemble FLIM and systems predictions and to establish how this is influenced by feed-back loops and perturbations. By testing these models on primary epithelial cells and tissues we will adapt the models iteratively to create a framework of response prediction for the network. Our ultimate goal is to derive probes and/or algorithms based on the models, suitable use with fixed or fresh human tissues to determine model robustness and network predictability in this real world setting.

Signal transduction by the ErbB receptor family

The ErbB receptor family are ubiquitously expressed growth factor-binding proteins that belong to subclass I of the superfamily of receptor tyrosine kinases (RTKs) [1]. This receptor family has four members (ErbB1-4); excessive signalling by these receptors upsets the balance between cell growth and apoptosis resulting in the development of a wide variety of solid tumours, including lung, mammary carcinoma, glioblastomas and squamous carcinoma [2]. In adulthood, insufficient signalling by this receptor family is linked to the development of neurodegenerative disorders, such as multiple sclerosis and Alzheimer's disease [reviewed in 3].

The receptors in the ErbB family have in common a structure (Fig. 1) consisting of an extracellular growth factor-binding domain (ectodomain), a single transmembrane α -helix, and a cytoplasmic domain containing a tyrosine kinase region, which is flanked by juxtamembrane (~40 residues) and C-terminal regulatory regions [1]. Upon binding their respective growth factors, the role of the ErbB receptor family is to transduce this signal across the plasma membrane by initiating

the catalytic activity of their intracellular kinase [3]. This is a poorly understood mechanism shown to be associated with the formation of almost every possible pair-wise combination of homo- and heterodimers (and perhaps larger oligomers) of the receptors in this family [4]. ErbB signals are terminated through the endocytosis of the receptor-ligand complex [5] (Fig. 1). Downstream intracellular signalling pathways are then initiated via the recruitment of a large repertoire of adaptors, enzymes and transcription factors, which activate the gene programmes that control cell proliferation, survival, adhesion, migration and differentiation [reviewed in ref.6].

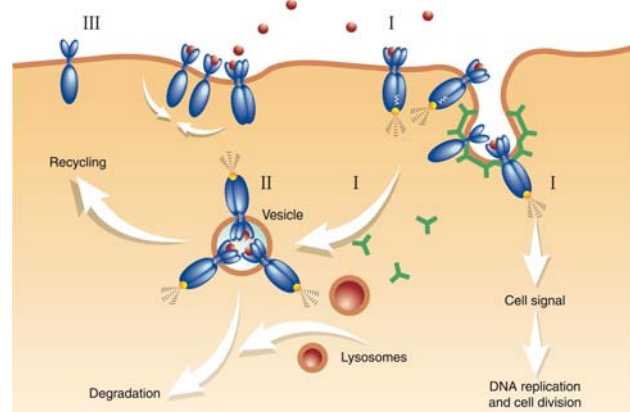


Figure 1. Schematic of receptor endocytosis. Receptors bind ligand at the cell surface (I) and are transported along the plasma membrane towards clathrin-coated pits to be internalised in vesicles (II). Then ligand-bound receptors are targeted for degradation in acidic intracellular compartments (lysosomes) or recycled to the cell surface. Endocytosis is used by the cell to downregulate receptor signals. It is common to find in cancerous cells the recycling pathway potentiated. [9]. All receptors in the EGFR family are endocytosed with the exception of ErbB2, which escapes endocytosis and, as a result, is the most oncogenic member of this receptor family.

Using single molecule fluorescence microscopy to study structure-function relationships in ErbB receptor signalling

Predictions from 3-D structures need to be tested and verified by experimental evidence from ErbB receptor molecules in the physiological environment of the cell. Unlike crystallised receptor fragments, intact receptors in cells are influenced by glycosylation, and by the plasma membrane's action potential, hydrophobic environments, 2-D geometrical restrictions and local curvature [7,8]. Interactions with other transmembrane receptors and/or intracellular substrates might also play an important role in the regulation of ErbB activation, as well as heterodimerisation, ligand-independent lateral propagation of activation via transient dimerisation and the formation of receptor oligomers [9,10]. These interactions are not explained by the structural data available and cannot be fully investigated either by ensemble fluorescence imaging technologies because these rely on non-physiological, and often ineffectual synchronisation of events, hence being inadequate to characterise conformational changes associated to signalling as the information can be lost in the ensemble average.

Single molecule fluorescence microscopy techniques can bridge the areas of structural biology and cell imaging by yielding constraints that have allowed us to derive *in situ* receptor structural information during signal transduction in cells [11]. To gain information on protein interactions and conformational changes it is necessary to resolve distances on the order of the size of the receptor. These can be accessed by using single pair FRET (spFRET). FRET is a phenomenon by which the excited-state energy of an optically excited molecule (donor) is transferred to a neighbouring molecule (acceptor) nonradiatively via intermolecular dipole coupling in the range 1-8 nm, a length-scale encompassing the typical diameter of membrane proteins [12]. Crucially, spFRET and single molecule fluorescence polarisation (smFP) can be measured simultaneously because the emission polarisation ratio of a single molecule is independent of the means of excitation, hence the acceptor/emission polarisation ratios can also be recorded after FRET excitation [13].

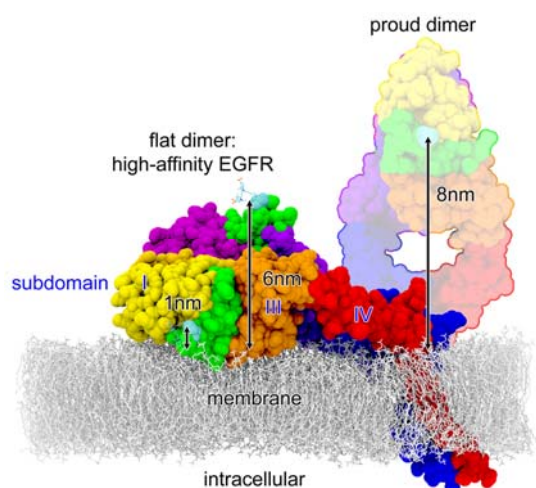


Figure 2. The back-to-back dimer of ErbB1 coloured by domain. The ErbB1 cognate ligand, the epidermal growth factor (EGF), is depicted in green. Subdomains I-IV of the ErbB1 receptor are numbered in the figure. The model is obtained from combining crystal structures as described in [14]. The figure shows the two possible orientations of the EGFR ectodomain dimer revealed by single molecule and FLIM-FRET microscopy. The two conformations are associated to the two different affinities shown by ErbB1 to its ligand.

The challenge of single molecule fluorescence imaging in mammalian cells

Single-molecule data on protein conformational changes in cell-free environments are much easier to quantify in the absence of out-of-focus cell autofluorescence. Total internal reflection (TIR) excitation allows the rejection of most, but by no means all out-of-focus background. This results in an unavoidable further reduction in signal-to-noise in the single molecule images from cells, which is even more acute when photons have to be split into different channels to collect spFRET and smFP data, where the signal is partitioned into four components discriminated in two colours and two polarisations to derive *in situ* protein structure [15]. In addition, as a side effect of TIR, fluorophores located at varied axial distances, as with proteins in the plasma membrane, experience different excitation field strengths. The fluorescence from single-molecules can therefore have a wider spread of signal-to-noise ratios than biomolecules immobilized on the same glass surface exposed to the same incident intensity. Using current image analysis technology we have successfully analysed data from those features bright enough to be easily distinguished from the background, but have had to discard significant amounts of potentially useful data.

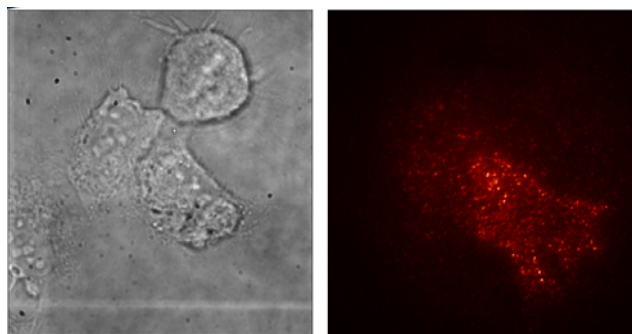


Figure 3. Wide field transmission (left) and single molecule fluorescence (right) images of HeLa cells. The EGFR molecules at the cell surface were labelled with EGF tagged with the red emitting fluorophore Atto647. The spots on the figure on the right show the location of single EGF/EGFR complexes in different oligomerisation stages. The number of fluorophores is determined by the number of photobleaching steps in each spot [15].

Using a Bayesian approach, pre-existing knowledge of the subject being measured and of the experimental apparatus used to make the measurements is explicitly included in the analysis, creating a much more powerful algorithm with trustworthy results where the limits of the analysis are clearly understood. This is in contrast to traditional ad-hoc methods where assumptions are often implicit and not appreciated by some who use them. Available prior knowledge has not yet been effectively included in the analysis of single molecule data, which in live cells leads to poor detection rates and crucially, to potentially erroneous conclusions. Additionally, every inferred result from a Bayesian analysis is accompanied by a confidence limit. This combination of increased detection rate and quantitative results make Bayesian techniques the optimal method to be applied to the analysis of spFRET and smFP data in live cells [16,17]

Ongoing experiments

We are currently choosing a preferred cell model(s) among a pool of ErbB1-4 expressing lung and breast cancer cell lines by screening their suitability to single molecule investigations. The best model should allow the study of heterocomplexes of all 4 receptors and mutant versions at single molecule level. We are also producing intracellular and extracellularly-tagged ErbB1-4 and cell lines expressing at least two receptor members to investigate combinatorial ErbB oligomerisation. We have begun

to use FRET to investigate ErbB1-ErbB2 interactions using fluorescent ligands and fused fluorescence proteins as probes which will be soon extended to ErbB2-ErbB3 complexes. We have also developed a method that enables single molecule nanoimaging in the X,Y plane of inter-molecular distances with <10 nm resolution in physiological conditions, bridging the gap between Forster Resonance Energy Transfer (FRET) and optical resolution. Its application allowed measurement of the much sought after distances between epidermal growth factor receptors predicted by dimer structures but too large for FRET. We are also investigating the effect of disrupting ErbB intracellular interactions with lipid rafts, actin, and microtubules on ErbB receptor oligomerisation and recruitment of SHC-GFP to the oligomers in HeLa cells. Finally, we are commissioning a 5-colour single molecule microscope to investigate the formation of macro-molecular complexes at the plasma membrane of living cells.

Conclusions

Pushing the frontiers of our understanding of signalling networks in the true physiological context requires a multidisciplinary approach. In this LSF programme we are utilising the sample preparation methods, fluorescence microscopy techniques and data analysis and simulation methods within the Octopus facility at the ensemble and single molecule levels to shed light on receptor structure-function relationships and interactions during signalling in cells. The ErbB receptor signalling network best exemplifies the significance of aberrations in the development of some of the most potent human malignancies. The further development of single molecule methods applicable to mammalian cells may reveal the alternative molecular mechanisms that ensure outputs in the de-regulated disease state, leading to a better understanding of positive and negative feed-back mechanisms used by cells to by-pass therapies.

Acknowledgements

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The role of BER proteins in the repair of DNA damage induced following NIR multiphoton laser microbeam irradiation

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Introduction

Deoxyribonucleic acid (DNA) is continually undergoing DNA damage and repair. DNA damage can arise from both endogenous and exogenous sources. Exogenous DNA damage occurs following exposure to ionising radiation. A specific feature of ionising radiation is the production of clustered DNA damage; when two or more lesions form within one to two helical turns of the DNA. It has previously been shown that these clustered lesions have reduced repairability compared to individual lesions (1, 2). Unrepaired DNA lesions can ultimately lead to chromosome aberrations mutations and possibly cancer therefore mammalian cells have developed highly conserved DNA repair pathways to repair damaged DNA.

The repair of damaged base lesions occurs predominantly via base excision repair (BER [Fig. 1]). Recognition of the damaged base occurs by a DNA glycosylase which cleaves the N-glycosidic bond to create an apurinic or abasic site (AP site). The DNA backbone is then cleaved by an AP endonuclease creating a single stranded nick 5' to the AP site (3). The repair can then progress through either short patch-BER (SP-BER) or long patch-BER (LP-BER) (Fig.1, (4)). In SP-BER, XRCC1 is recruited rapidly to the nick and acts as a scaffold protein for polymerase β (Pol β) and Ligase III (5). Pol β inserts the missing base and cleaves the 5' phosphate to allow ligation by Ligase III. When Pol β is unable to cleave the 5' phosphate, BER proceeds by LP-BER. During LP-BER, Pol δ or Pol ϵ is recruited to insert bases in a proliferating cellular nuclear antigen (PCNA) dependent manner, which results in DNA strand displacement (Fig. 1 (3)). Flap structure specific endonuclease 1 (FEN1) cleaves the DNA flap to create a ligatable structure before ligation occurs by Ligase 1 (3).

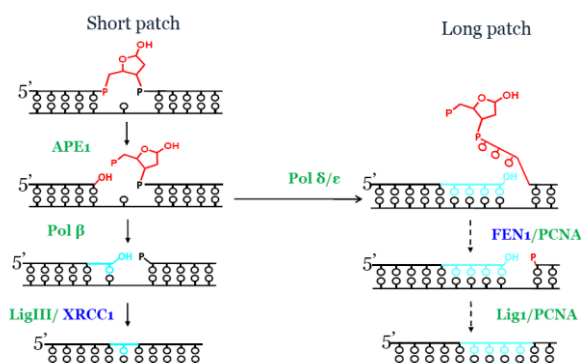


Figure 1: Schematic diagram representing the repair of base lesions by BER.

Previously we have shown that NIR laser microbeam irradiation induces DNA double strand breaks of different complexity (6, 7). Our initial studies demonstrate that following irradiation, base lesions are induced within the DNA and recruit proteins involved in both SP-BER and LP-BER (represented by the recruitment and loss of XRCC1-YFP and FEN1-GFP respectively). Clustered DNA damage is known to be repaired in part by LP-BER. We interpret the different repair kinetics in terms of the repair of single strand breaks (SSBs) and SSBs in clustered damage sites processed by SP- and LP-BER.

Methods

Cell culture

EMC11 (XRCC1^{-/-}) cells were cultured in Dulbecco's modified minimum essential medium (DMEM) supplemented with 10% foetal calf serum (FCS), 100 μ g/ml penicillin streptomycin and 2mg/ml L-glutamine at 37°C with 5% CO₂ in air.

Transient transfection of EMC11 cells

EMC11 cells were plated at 2×10^5 cells per 30 mm glass bottom dishes 24 h prior to transfection. Cells were transfected with 5 μ g of XRCC1-YFP (a gift from Dr. G. Dianov) or FEN1-GFP (Insight Biotechnology, UK) DNA according to the Qiagen SuperFect[®] transfection reagent protocol. In brief, 5 μ g of DNA was added to a total volume of 100 μ l OptiMEM. SuperFect[®] transfection reagent (15 μ l) was added for 5-10 min followed by the addition of 600 μ l of complete culture medium. The transfection reagent was added to the cells and incubated for 3 h. The transfection reagent was removed by washing 3 x in 1 ml phosphate buffer saline and cells were incubated for 24 h prior to irradiation.

Laser set-up

A Ti:Sa laser (Mira 900, Coherent Inc., USA) tuned to 730 nm was focused through a x 60 water microscope objective, numerical aperture 1.20 of an inverted microscope (TE2000, Nikon). A customised computer software programme (LabView[™]) operated the automated microscope stage in the x- and y-plane at a step size of 16 μ m.

Real time NIR multiphoton laser microbeam irradiations.

Cells were incubated with 10 μ g/ml Hoechst dye for 10 min prior to irradiation at 37°C. A red bandpass filter (RG610) was placed on top of the culture dish to prevent the Hoechst dye from absorbing UV light from ambient light. Cells were maintained at 37°C throughout the irradiation using the temperature control chamber connected to a pumped water circulator (Neslab RTE7 Digital One, Thermo Scientific). The laser was set to a wavelength of 730 nm and a nominal power of 10 mW measured through the x 40 air, numerical aperture

0.95, microscope objective. Cells were irradiated in culture medium using a x 60 water objective. Time 0 was recorded immediately following irradiation of the cells (less than 10 s) and images were collected at the stated times following irradiation using confocal microscopy (EC1, Nikon) equipped with an argon ion laser at 488 nm. The images were collected using three Kalman filtered scans with a minimum of 10 cells visualised for analysis per experiment.

Results

Real time kinetics of recruitment and loss of XRCC1-YFP to DNA damage induced by NIR multiphoton laser microbeam irradiation

Cycling EMC11 cells transiently transfected with XRCC1-YFP were irradiated using the NIR laser microbeam to determine the real time recruitment and loss of XRCC1-YFP at sites of DNA damage. XRCC1-YFP is rapidly recruited to sites of DNA damage reaching maximal levels within 2-3 min (Fig. 1 inset). The level of XRCC1-YFP fluorescence is 30% of the maximal fluorescence at 60 min following irradiation. At 240 min, a residual relative fluorescence level of 15% was determined. The repair of NIR laser microbeam induced lesions occurs with bi-exponential decay kinetics. The majority of repair (70%) occurs via a fast component with a half-life of 14 ± 6 min. A small component (30%) is repaired with a significantly longer half-life of 230 ± 175 min.

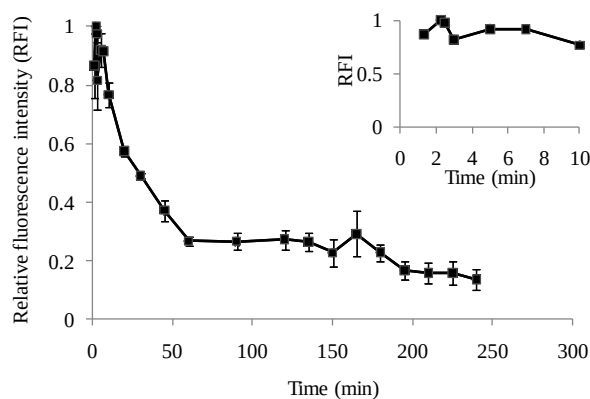


Figure 2: Real time recruitment and loss of XRCC1-YFP fluorescence to sites of DNA damage following NIR multiphoton laser microbeam irradiation. The inset represents the data expanded to show the first 10 min in more detail. Each point represents the normalised relative fluorescence intensity of a minimum of 10 cells per experiment and the mean of 3 independent experiments $\pm$ SEM.

Real time kinetics of recruitment and loss of FEN1-GFP following NIR laser microbeam irradiation

Cycling EMC11 cells were transiently transfected with FEN1-GFP and irradiated using the NIR multiphoton laser microbeam. Following irradiation, FEN1-GFP is rapidly recruited to sites of induced DNA damage. Maximal relative fluorescence was seen later than that of XRCC1-YFP (~ 2 min) at ~ 5 min following irradiation (Fig. 3 inset). Within 15 min, FEN1-GFP relative fluorescence intensity is 15% of the maximum fluorescence. FEN1-GFP reaches background fluorescence intensity levels within 60 min following irradiation (Fig. 3). The repair of induced lesions involving FEN1-GFP is readily fitted by a single exponential decay with a half-life of 15 ± 2 min. this value is similar to that seen with XRCC1-YFP.

Discussion

The data presented show that following NIR multiphoton laser microbeam irradiation key proteins involved in SP-BER

(XRCC1-YFP) and LP-BER (FEN1-GFP) are recruited to sites of induced DNA damage. XRCC1-YFP is involved in the repair of NIR multiphoton laser microbeam induced damage with bi-exponential repair kinetics suggesting that repair occurs by two distinct processes. It is suggested that the fast component of repair represents repair of less complex DNA damage which is consistent with SP-BER repair of simple DNA lesions (Fig. 4). This is consistent with previous investigations by Dikomey and Franzke (8) that showed that DNA strand break repair occurs with three distinct half-lives (Table 1). The fast

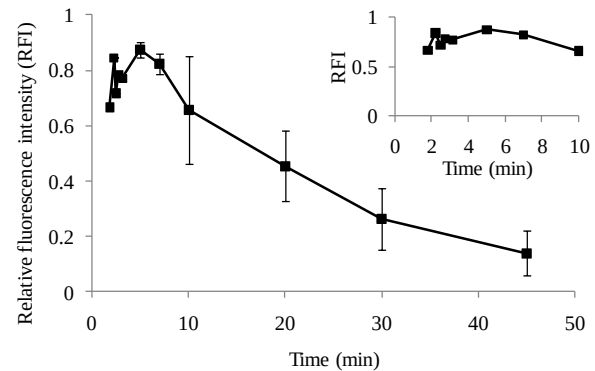


Figure 3: Real time recruitment and loss of FEN1-GFP fluorescence following NIR multiphoton laser microbeam irradiation. The inset represents the data expanded to show the first 10 min in more detail. Each point represents the normalised relative fluorescence intensity of a minimum of 10 cells per experiment and the mean of 3 independent experiments $\pm$ SEM.

component of strand break repair (2 min) suggesting that NIR multiphoton laser microbeam irradiation may not induce these very simple damages or XRCC1 may not be required for the repair of this sub-set of damage. Following laser microbeam irradiation, the half-life of the component of repair involving XRCC1 is consistent with the 2nd half-life observed for strand break repair (Table 1). The slow component of repair involving XRCC1-YFP may represent the repair of complex DNA damage that is more difficult to repair than simple damage (Fig. 4) or alternatively may represent the repair of DNA DSBs by an back-up non-homologous end joining pathway that has been proposed to involve XRCC1 (Fig. 4). DSBs would have slower repair kinetics than the DNA lesions. The slow kinetics of XRCC1-YFP is similar to the slow component of strand break repair is shown by Dikomey and Franzke (~ 200 min, Table 1, (8)).

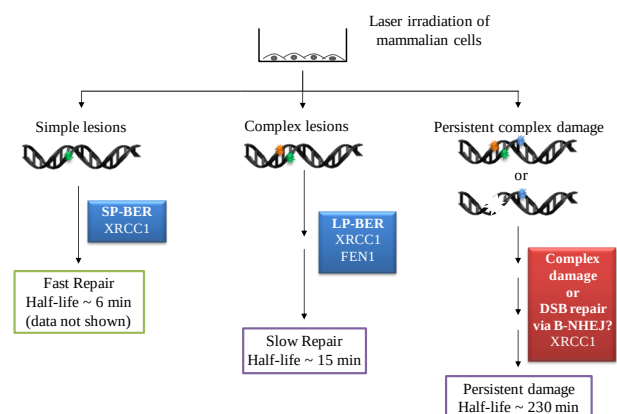


Figure 4: Schematic diagram illustrating the induction and repair of DNA damage following laser microbeam induced damage.

FEN1-GFP is recruited more slowly to induced DNA damage than XRCC1 (~ 5 min vs. ~ 2 min, respectively). The role of

FEN1 in LP-BER (Fig. 1) has been shown to be the predominant repair pathway for repair of complex DNA lesions including complex SSBs. Repair involving FEN1-GFP occurs with single exponential decay kinetics suggesting repair is by a single process consistent with the second component of DNA strand break repair (~15 min [Fig. 4 and Table 1 (8)]). The similarity of the loss of XRCC1-YFP and FEN1-GFP may indicate that XRCC1 is involved in LP-BER. The lack of significant slow process with FEN1-GFP would be consistent with the slow loss of XRCC1-YFP results for repair of DSBs or some other form of damage which does not require BER.

	1 st Half-Life (% relative contribution)	2 nd Half-Life (% relative contribution)	3 rd Half-Life (% relative contribution)
DNA strand break repair (Dikomey & Franzke, 1986)	2 min (70%)	17 min (25%)	200 min (5%)
XRCC1-YFP		NIR laser microbeam 15 min (70%)	NIR laser microbeam 230 min (30%)
FEN1-GFP		NIR laser microbeam 15 min	

Table 1: Comparison of kinetic half-lives of BER proteins at induced DNA damage compared to those of DNA strand breaks (8).

Conclusions

It is concluded that sub-sets of NIR laser microbeam induced DNA damage are repaired by XRCC1 and FEN1. XRCC1 and FEN1 repair DNA damage with a half-life of ~15 min which may represent processing of complex damage by BER. XRCC1 although not FEN1 is involved in the repair of a subset of DNA damage with long repair kinetics (~230 min) which may represent very complex DNA damage of DNA or DSBs which are not repaired by BER.

Acknowledgements

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Time Resolved Infrared Analysis of AppA_{BLUF} using Isotopic Labeling at Mutant Studies

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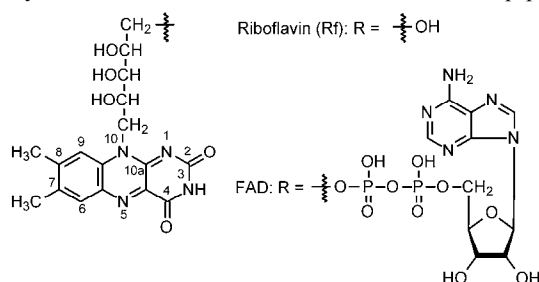
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Introduction

Blue light using FAD (BLUF) domain proteins exist in many species with a variety of functions [1-5]. BLUF domain proteins bind flavin noncovalently in order to sense and respond to the intensity of blue (450 nm) light. AppA is the first and best characterized BLUF domain photoreceptor and is found in *Rhodobacter sphaeroides* where it acts as an antirepressor of photosystem biosynthesis. AppA consists of two domains: the N-terminal BLUF domain and a C-terminal domain that is responsible for the oxygen sensitivity of the protein. Our initial studies have focused on a truncated form of AppA that consists only of the N-terminal domain (AppA_{BLUF}). Initial studies of the BLUF domain identified early structural changes in photoactivity [1]. These studies determined the formation of a signaling state which is characterized by a 10 nm red shift in $\lambda_{\max}$ of the flavin chromophore. Currently, the goal is to understand the underlying chemical structures of the dark and signaling state of AppA by using spectroscopic analysis.

Isotope Labeling of the Flavin Chromophore

The normal chromophore in AppA is flavin adenine dinucleotide (FAD). Since isotope labeling of the isoalloxazine ring in FAD requires first synthesis of the ring and then biosynthetic conversion to FAD, which is a multistep process,



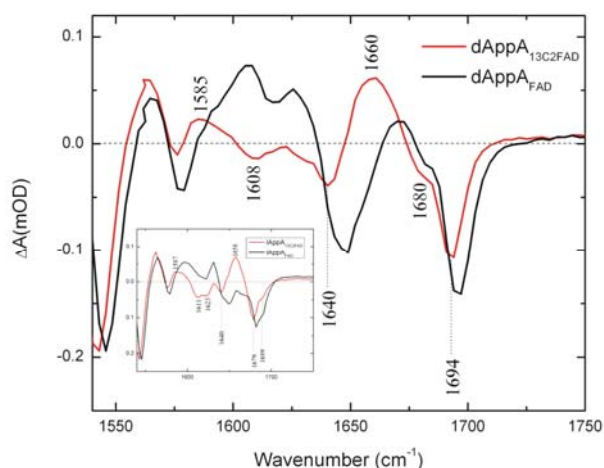


Figure 2: TRIR Spectra of AppA_{BLUF} Bound to Unlabeled FAD and [2-¹³C₁]FAD.

¹³C labelling on the IR spectra of FAD in H₂O and D₂O. We have established that deuteration at N3 (N3-H to N3-D) alters the coupling between the C2=O and C4=O groups in the isoalloxazine ring. Since coupling is affected by deuteration, the effect of ¹³C labelling will thus depend on whether spectra are collected in H₂O or D₂O. Our plans are to obtain TRIR data of AppA_{BLUF} in H₂O buffer but this is a very challenging experiment since short path length cells must be used (6 μm instead of the normal 50 μm).

Isotope Labeling of the AppA Protein

The 1669 cm⁻¹ transient observed in the TRIR spectrum of dAppA_{BLUF} has been described previously as a photoactive marker and is only seen in AppA_{BLUF} mutants that undergo a photocycle. This mode has previously been assigned to the amide side chain of Q63 which is thought to rotate or undergo keto-enol tautomerism upon photoexcitation. We have expended a very significant effort in attempts to isotopically label this glutamine in AppA_{BLUF}. This has included generating a variant of AppA_{BLUF} (6Q-N) in which all the glutamines apart from Q63 (6) have been replaced with asparagines. Remarkably the 6Q-N AppA_{BLUF} has a photocycle indistinguishable from the wild-type protein. However we have so far been unsuccessful at selectively labeling Q63 in 6Q-N AppA_{BLUF}. This has been attempted by growing cells expressing this protein on labeled glutamine and using a transaminase-deficient cell line to reduce scrambling of label. Currently, yields of protein are too low to permit the analysis required to ensure isotope incorporation.

While we attempt to improve the yield of the 6Q-N AppA_{BLUF} protein, we have expressed the wild-type protein on minimal media containing ¹⁵NH₄Cl so that all the nitrogens in the protein are labeled with ¹⁵N. This approach will allow us to identify protein modes in the TRIR difference spectra. In particular, if the 1669 cm⁻¹ transient is due to the Q63 amide side chain, then this mode should be sensitive to ¹⁵N labeling. Uniformly labeled AppA_{BLUF} will be studied during our next RAL trip.

Mutants of AppA_{BLUF}

We have generated the following AppA_{BLUF} mutants: W104A, M106F, M106A, W104A/M106F, S41T, Q63E, Y21S and Y21C. These amino acids are distributed around the isoalloxazine ring (**Figure 3**) and all have been proposed to play key roles in the AppA photocycle. We will not describe in detail here all the results of each mutation, the Q63E AppA_{BLUF} is extremely interesting. Previous models of AppA excitation incorporated changes in hydrogen bonding interactions involving Q63. Several mutants have been prepared of Q63, all

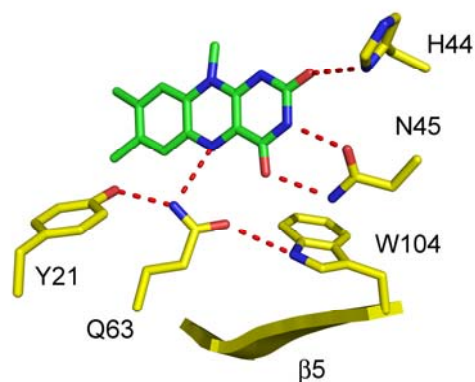


Figure 3: The Isoalloxazine Binding Pocket in AppA.

of which are photoinactive (such as Q63L) and that usually mimic the light adapted form of the protein. Q63E in contrast has spectroscopic properties that are intermediate between light and dark states. The TRIR spectra together with time resolved fluorescence and transient absorption measurements all indicate a unique protein-chromophore complex that, when fully characterized, should shed light on the structural changes that accompany photoactivation. Initial TRIR spectra are shown in **Figure 4** where it can be seen that major differences exist in the modes attributed to the flavin C=O groups. The 1650 cm⁻¹ peak has split into two bands, while the highest wavenumber C4=O mode is shifted to 1720 cm⁻¹, consistent with reduced hydrogen bonding in the mutant. In addition, [4-¹⁸O₁]FAD will be incorporated into this mutant to assign this mode the C4 carbonyl of FAD.

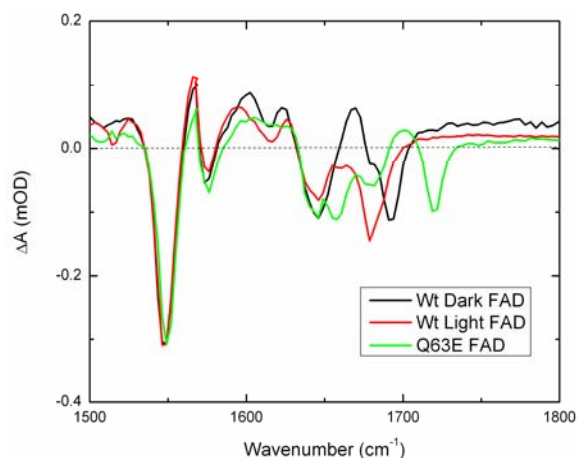


Figure 4: TRIR Spectra of Q63E AppA_{BLUF}
TRIR spectra (2mM, 3ps) are shown of dAppA_{BLUF} (black), lAppA_{BLUF} (red) and Q63E (green).

Conclusions

Both AppA's dark and light excited state dynamics have been studied using time resolved infrared spectroscopy (TRIR). TRIR spectra of unbound flavin were obtained and used to determine ground state and excited state modes. In addition, TRIR spectra of dark AppA and light AppA were also obtained and the differences between unbound flavin modes and those modes that correlated to protein or protein-flavin interactions were assigned. These studies showed some significant differences between the flavin, light and dark AppA [6].

We have used isotope labeling of both the flavin chromophore and of the protein in order to identify major differences in the TRIR spectra of light AppA and dark AppA. The data gathered allowed for the assignments of the C4 and C2 carbonyl modes that shift to lower frequency upon photoconversion to lAppA. In addition, we have studies

structurally conservative mutants of AppA using TRIR. These mutants, such as Q63E, shed light upon the sensitivity of the hydrogen bonding network of protein residues to the flavin.

Future work will include unnatural amino acid incorporation into position Y21 which has the potential to alter the kinetics of the photocycle by either eliminating or enabling electron transfer. Fluorotyrosines with a reduced pKa values compared to tyrosine have been introduced to Y21 and are thus expected to be valuable probes of the electron transfer reaction in which Y21 is thought to participate. TRIR measurements will give important information on how the kinetics are effected with the incorporation of various fluorinated tyrosines.

Acknowledgements

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High-resolution stimulated Raman spectroscopy with photoacoustic detection (PARS) of formic acid dimer

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Introduction

Spectroscopy is a powerful tool to probe intermolecular bonding. Raman and IR spectroscopy are complementary techniques, employing different selection rules. Quantum chemical calculations can be used in addition to spectroscopy to provide a deeper insight into bonding environments.

Hydrogen bonding $X-H\cdots Y$, is an important intermolecular interaction. Usually a weakening of the X-H bond is involved, and hence a shift towards lower wavenumber ('red shift') of the XH-stretching vibration. Many recent works have shown evidence of a hydrogen bond which shows a strengthening of the X-H (e.g. C-H) bond and hence a shift towards higher wavenumber ('blue shift') in the XH-stretching vibration.

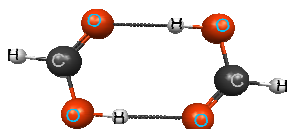


Fig. 1: CP-corrected MP2/6-311G(d,p)++ geometry of formic acid cyclic dimer.

The lowest energy dimer conformation of formic acid (the cyclic dimer) has an energy low enough to be abundant and easily observable at room temperature in the gas phase. This cyclic dimer has predicted vibrational shifts towards lower (OH-stretching vibrations) and higher wavenumber (CH-stretching vibrations) and thus provides a unique opportunity to investigate the origin of vibrational shifts in more detail.

Raman spectroscopy can access the symmetric stretching vibrations, and infrared spectroscopy antisymmetric stretching vibrations. Conventional Raman experiments, however, suffer from low sensitivity and spectral resolution. More advanced techniques are therefore required to record Raman spectra of gas phase species with high sensitivity and sufficient spectral resolution.

High-resolution stimulated Raman spectroscopy with photoacoustic detection (PARS)

A stimulated Raman experiment with photoacoustic signal detection has been set up, which allows measuring Raman spectra of gaseous species with high resolution and sensitivity [1]. In the experiment, 532 nm laser light (second harmonic of a seeded Nd:YAG laser) is the Raman pump beam which is combined with the tuneable output around 630 nm of a dye laser as the stimulating beam and focused into a cell (EPSRC Laser Loan Pool, NSL1). If the energy difference between the laser photons corresponds to an allowed Raman transition, molecules are promoted to a vibrationally excited state by the stimulated Raman process. By collisions, the vibrational excitation is converted into local heating which creates a pressure wave which is picked up by a microphone (photoacoustic Raman spectroscopy, PARS, [1,2]). Since the seeded Nd:YAG laser has a very narrow bandwidth, the resolution of stimulated Raman scattering is only limited by the bandwidth of the stimulating dye laser (ca. 0.05 cm^{-1}).

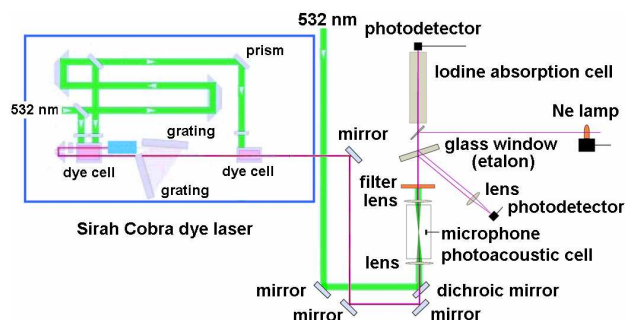


Fig. 2: Schematic of stimulated Raman photoacoustic setup.

A system enabling accurate wavelength calibration of the tuneable dye laser has been set up and characterised, encompassing monitoring etalon fringes from a glass window, optogalvanic Ne spectroscopy, and monitoring iodine absorptions in a heated iodine vapour cell, achieving 0.03 cm^{-1} accuracy within a 20 nm scan range [1,3,4].

Extension of PARS to measure corrosive gases

The microphone inside the photoacoustic cell cannot sustain harsh environments like corrosive gas samples, as would be the case for measuring formic acid in the gas phase. We extended the PARS technique to the measurement of corrosive gases and hostile environments using a modification of the 'optophone' detection of Zare and coworkers [5]. In this approach, the pressure wave induced by light absorption distorts a membrane which has been gold coated to be corrosion resistant. The distortion is detected by a laser pointer and a split photodiode outside of the photoacoustic cell.

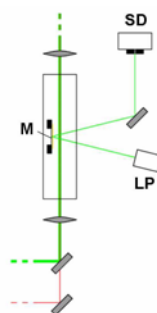


Fig. 3: 'Optophone' photoacoustic detection. M: gold-coated membrane; LP: laser pointer; SD: split photodiode.

CH-stretching vibration of formic acid dimer

High-level quantum chemical calculations were performed to support the experiments, using the Gamess *ab initio* programme package with MP2 correction for electron correlations and the triple-zeta 6-311G(d,p)++ basis functions, and including a self-written program implementing Counterpoise Correction (CP) for the Basis Set Superposition Error (BSSE) [6]. The calculations predict a blue shift of 30 cm^{-1} for the Raman-active symmetric and 32 cm^{-1} for the IR-active antisymmetric CH stretching vibration upon dimerisation.

Stimulated Raman spectra have been acquired for gaseous formic acid, using the PARS technique described above with optophone detection of photoacoustic signals. The CH-stretching vibration of both monomer and dimer formic acid has been observed (see fig. 4). The Raman-active symmetric CH-stretching vibration exhibits a shift towards higher wavenumber ('blue shift') of 7 cm^{-1} at room temperature.

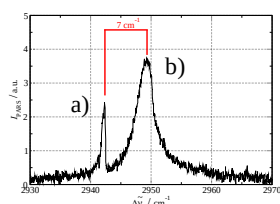


Fig. 4: Stimulated Raman photoacoustic spectrum (PARS) of formic acid (49 mbar) at room temperature. (a) Monomer, (b) dimer.

The IR-active antisymmetric stretching vibration has also been observed to investigate whether the vibrational shift can be partly attributed to the splitting between the symmetric and antisymmetric stretching vibration modes. FTIR spectra were obtained using a Perkin Elmer Spectrum 6X FTIR Serial 50895 machine, with a resolution of 0.2 cm^{-1} . Spectra obtained at high pressures ($\sim 20\text{ mbar}$) will contain predominantly dimers, and at low pressures ($\sim 2\text{ mbar}$) predominantly monomers. Suitable subtraction gives pure monomer and dimer spectra (see fig. 5).

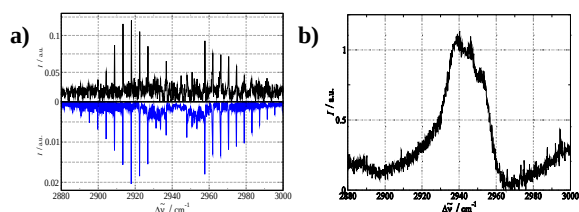


Fig. 5: (a) FTIR spectrum of gaseous formic acid monomer ($\sim 1.2\text{ mbar}$) (top) and PGOPHER simulation (bottom). (b) FTIR spectrum of gaseous formic acid dimer ($\sim 18.6\text{ mbar}$).

The monomer spectrum is well reproduced by a simulation using the PGOPHER programme [7]. IR spectroscopy shows a shift of ca. 4 cm^{-1} towards higher wavenumber ('blue shift') for the antisymmetric CH-stretching mode at room temperature.

The shift of the CH-stretching vibration towards higher wavenumber is thus genuine and not due to the splitting of the symmetric/antisymmetric modes. The shift is in qualitative agreement with the calculations, but smaller than calculated. This is most likely an effect due to thermal excitation; at room temperature, many low-frequency intermolecular vibrational modes are expected to be populated. This significantly weakens the hydrogen bond, and therefore the frequency shift induced by hydrogen bonding as a secondary effect is less pronounced. This effect shows clearly that theoretical calculations on weak hydrogen bonds which do not include thermal excitation can differ significantly from experimental observations at room temperatures (see also ref. [6]).

Mechanism of the vibrational frequency shift

Significant vibrational shifts are usually being associated with a bonding environment and have been used in the past as a signature of hydrogen bonding. The observed shift is thus unexpected and somewhat counter-intuitive at first, since the CH group is *not* involved in the hydrogen bonding of the cyclic dimer (see fig. 1). We propose that the shift is due to a secondary effect of the hydrogen bonding between the C=O and the O-H in cyclic formic acid dimer, based on our *ab initio* calculations.

The electric dipole moment of a bond is defined as partial charge multiplied by distance. If partial charges remain constant, then with increasing bond length, the dipole moment will increase. Our calculations show, however that for the C-H bond of formic acid the dipole moment will decrease with

increasing bond length since partial charges of C and H are becoming weaker. This means that an increase in partial charges (polarity) in the C-H bond will lead to a compression and strengthening of the bond, causing a shift towards higher wavenumber ('blue shift') for the stretching vibration. The increase in polarity could be due to a secondary effect: upon formation of the hydrogen bond between O-H and C=O, the C=O bond becomes more polar which causes a withdrawing effect through the molecule, increasing the polarity and the strength of the CH group.

This mechanism will also apply for open dimer structures, and it will contribute significantly to the blue shift of the CH-stretching vibration predicted by *ab initio* calculations for these structures. The lowest energy open configuration of formic acid dimer, e.g., has a strong OH $\cdots$ O bond, and a weaker CH $\cdots$ O hydrogen bond (see fig. 6).

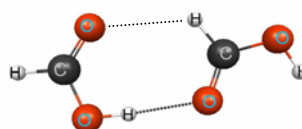


Fig. 6: CP corrected, MP2/6-311G(d,p)++ geometry of the lowest energy open configuration of acid dimer.

Conclusions

Using a laser loan from the EPSRC Laser Loan Pool, we have successfully set up a stimulated laser Raman experiment with optophone photoacoustic detection which allows the observation of high-resolution Raman spectra of gas-phase species in hostile environments. With this technique, we have observed the CH-stretching vibration of formic acid dimer and found a distinct shift of this vibration towards higher wavenumber ('blue shift'). This shift is not directly due to intermolecular hydrogen bonding in the cyclic dimer. We propose that it is due to a secondary effect of the hydrogen bonding of the OH with C=O, where increase of electron density to the C=O bond by hydrogen bonding increases the polarity and strength of the C-H bond, thus inducing a shift towards higher wavenumber.

In future work, we would also like to apply this experimental technique to investigate the Raman-active symmetric OH-stretching vibration of cyclic formic acid dimer, for example to study the concerted double-proton transfer in the two equivalent OH $\cdots$ O hydrogen bonds in the nanosecond time domain.

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Transient 2D-IR Spectroscopy of [FeFe]Hydrogenase Enzyme Model Compounds

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Introduction

The [FeFe]hydrogenases are a family of metalloenzymes with an active site based upon a $2\text{Fe}_2\text{S}$ cluster in which the Fe atoms are coordinated by a mixture of carbonyl and cyano ligands. Interest in these systems is driven by the fact that they are capable of catalyzing the activation of molecular hydrogen. Furthermore, it has been shown that model compounds of the form $(\mu\text{SRS})\text{Fe}_2(\text{CO})_6$ are also capable of performing this function leading to the belief that these materials may form the basis for the next generation of catalytic materials for hydrogen fuel cells.¹⁻⁴

In order to maximize the potential of these model systems in technological applications, it is imperative that we obtain a thorough understanding of their solution-phase chemistry and dynamics. One particularly relevant facet of their chemistry relates to the production of a vacant coordination site at one of the Fe centres. This is inherent in the active form of the enzyme and can be produced in solution by the photolytic removal of a carbonyl ligand.⁵ Ultrafast infrared spectroscopy techniques have been shown to provide an effective probe of this process by virtue of the fact that carbonyl ligands give rise to structurally sensitive patterns of intense absorption bands in the spectrally uncongested region of the mid-IR at wavelengths near $5\text{ }\mu\text{m}$. Recently, we have employed transient infrared pump-probe (TRIR) spectroscopy to study the carbonyl ligand photolysis of $(\mu\text{S}(\text{CH}_2)_3\text{S})\text{Fe}_2(\text{CO})_6$ (**1**) at 350 nm in heptane solution. This showed the formation of a complex set of photoproduct absorption bands consistent with CO ligand removal, but the linear nature of the data precluded definitive assignment of the peaks to a single photoproduct or mixture of species.⁶ Ultrafast 2D-IR spectroscopy is a useful tool for problems such as this due to the ability to spread the molecular response over two frequency dimensions and thus give access to previously inaccessible information.^{7,8} Applications of transient 2D-IR (T-2D-IR) spectroscopy to the photolysis of **1** in heptane have subsequently been used to determine both the vibrational coupling patterns of the photoproduct absorptions via the presence of off-diagonal peaks in the 2D spectrum and the relative appearance rates of these peaks following photolysis in order to assign the bands to a single photoproduct species.⁹ Furthermore, these experiments observed a more rapid vibrational relaxation rate of the photoproduct in comparison to that of the parent,¹⁰ allowing this single photoproduct to be assigned to a solvent adduct species in which the $\text{Fe}_2(\text{CO})_5$ cluster was coordinated by a heptane molecule at the vacant site. Finally, it has been shown by ns-TRIR that photolysis of **1** in a mixed solvent consisting of a cyanoheptane in a molar majority of heptane leads to rapid formation of the heptane adduct from the first solvation shell followed by competitive substitution of heptane by the more strongly-coordinating solvent species.¹¹

Here, we employ transient 2D-IR (T-2D-IR) spectroscopy to investigate the vibrational coupling patterns of the photolysis products of **1** in neat

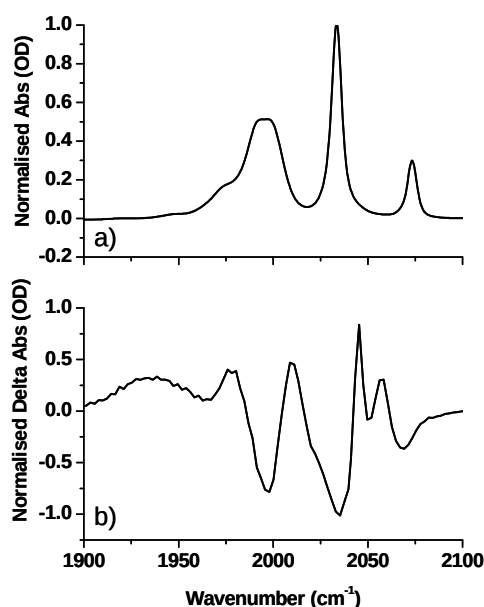


Figure 1: a) FTIR spectrum of $\sim 1\text{mM}$ solution of **1** in cyanoheptane solution. b) TRIR spectrum of the same solution following photolysis at 350 nm . The data shown corresponds to a $\text{UV}_{\text{pump}}\text{-IR}_{\text{probe}}$ delay time of 200 ps .

cyanoheptane in order to determine whether these too are consistent with the formation of a single solvent adduct photoproduct.

Experimental

The T-2D-IR spectrometer has been described in detail elsewhere.¹² Briefly, the ULTRA laser system was used to generate mid IR pump and probe pulses resonant with the CO stretching vibrations of **1** via optical parametric amplifiers (OPA) equipped with difference frequency mixing of the signal and idler beams. For 2D-IR spectroscopy, the narrow bandwidth ($\sim 10\text{ cm}^{-1}$) pump pulse (IR_{pump}) was generated by an OPA pumped by the ps pulse duration arm of the twin synchronized regeneratively amplified Ti:sapphire laser systems, while the broad bandwidth ($\sim 400\text{ cm}^{-1}$) IR_{probe} pulses originated from a second OPA pumped by the fs pulse duration regenerative amplifier. For T-2D-IR measurements, the UV photolysis pulse (UV_{pump}) was produced by a third OPA, pumped by the fs-regenerative amplifier and equipped with fourth harmonic generation capability in order to reach the required 350 nm wavelength.

The data presented here were recorded by modulating the amplitude of the two pump pulse trains (UV_{pump} and IR_{pump}) at one quarter and one half of the laser pulse repetition rate (10 kHz) respectively. This enabled TRIR, 2D-IR and T-2D-IR data

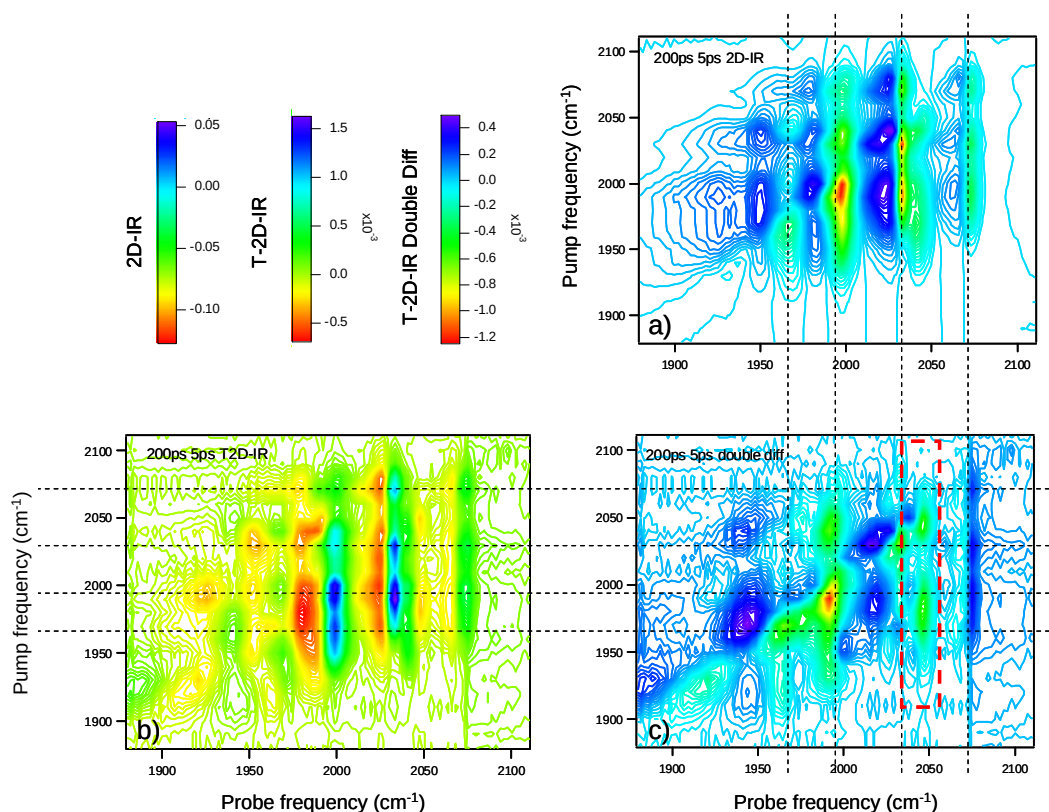


Figure 2: a) 2D-IR spectrum of **1** in cyanoheptane solution recorded with 5 ps $\text{IR}_{\text{pump}}\text{-IR}_{\text{probe}}$ delay time and magic angle polarization relationship between IR_{pump} and IR_{probe} pulses. b) T-2D-IR data with 200 ps $\text{UV}_{\text{pump}}\text{-IR}_{\text{pump}}$ delay time, 5 ps $\text{IR}_{\text{pump}}\text{-IR}_{\text{probe}}$ delay time and parallel polarization relationship between UV_{pump} and IR_{probe} pulses and magic angle relationship between IR_{pump} and IR_{probe} pulses. c) Double difference T-2D-IR spectrum in which the data in a) has been scaled and subtracted from that in b) to reveal only peaks due to the photoproduct.

to be collected in sequential laser pulses along with a background signal (no pump pulses present).

The samples were held between CaF_2 windows separated by a PTFE spacer of 100 μm thickness. The solute **1** was produced and handled via established methods¹ and all solvents were obtained from Sigma Aldrich and used without further purification. To prevent sample degradation and window damage, the solution was flowed and the sample cell rastered in a plane perpendicular to the laser beam direction.

Results and Discussion

Figure 1 shows the FTIR spectrum and TRIR data for **1** in cyanoheptane solution. The FTIR spectrum (Fig 1(a)) consists of three intense transitions in the carbonyl stretching region, located at 2073, 2033 and 1997 cm^{-1} . These are very close to the absorption peaks observed for **1** in heptane solution,¹⁰ though the significant broadening caused by the polar solvent leads to reduced spectral resolution to the point where it can only just be determined that the 1997 cm^{-1} peak consists of three overlapping transitions.

Upon photolysis at 350 nm with a $\text{UV}_{\text{pump}}\text{-IR}_{\text{probe}}$ time delay of 200 ps, the TRIR spectrum of **1** (Fig 1(b)) shows three strong bleaches due to the loss of population of the parent molecules alongside positive features due to the formation either of hot parent molecules or photoproducts. The positive peaks are located at 2056, 2045, 2008, 1975 and 1930 cm^{-1} respectively though it is non-trivial to differentiate between them in terms of assignment via linear spectroscopy alone.

In order to assign these features more definitively, T-2D-IR spectra were obtained with a $\text{UV}_{\text{pump}}\text{-IR}_{\text{pump}}$ time delay of 200 ps and an $\text{IR}_{\text{pump}}\text{-IR}_{\text{probe}}$ time delay of 5 ps. The former was used to select a dynamically quiet region of the spectrum where the

population of the photoproduct was, according to TRIR studies,¹¹ not changing dramatically, while the latter was chosen to maximize the signal strength of the T-2D-IR peaks and minimize coherence effects due to overlap between the IR_{pump} and IR_{probe} pulses. The results of these experiments are shown in Figure 2. Fig 2(a) shows the 2D-IR spectrum data channel, which should only show peaks due to the parent molecule. This is indeed the case, the negative peaks on the diagonal located at $(\omega_{\text{probe}}, \omega_{\text{pump}}) = (2073, 2073)$, $(2033, 2033)$ and $(1997, 1997)$ correspond well to the $\nu=0\text{-}1$ transitions of the parent molecule while a peak located at $(1975, 1975)$ is due to the shoulder observed in the FTIR spectrum at this frequency but which is better-resolved in the 2D-IR data by virtue of spreading the response over two axes. In each case, these diagonal features are accompanied by positive peaks shifted by 5-10 cm^{-1} to lower probe frequency that correspond to the $\nu=1\text{-}2$ transitions of the diagonal modes; the wavenumber shift is caused by the anharmonicity of the vibrational potential functions. Finally, each diagonal peak is linked to each of the other three by off-diagonal peak pairs that indicate vibrational coupling between the CO stretching modes. This is consistent with previous observations of the 2D-IR spectroscopy of **1** in organic solvents.¹⁰

These same features are present in the T-2D-IR channel (Fig 2(b)), though it is noted that the phase of these signals are reversed. By this, it is meant that the $\nu=0\text{-}1$ contributions become positive peaks while the red-shifted $\nu=1\text{-}2$ features are negative-going. This is caused by the presence of the UV_{pump} pulse in the T-2D-IR measurements leading to a reduced population of the parent molecule and a subsequent bleach of the 2D-IR peaks due to this species in the T-2D-IR spectrum. The difference-spectrum representation of the T-2D-IR spectrum means that these reversed phase features are the 2D equivalent of the bleaches observed in the TRIR data (Fig 1(b)).

Comparison of the T-2D-IR data with the 2D-IR channel also shows a significant number of additional features that are assignable to the 2D-IR spectrum of the photoproduct species. In particular these are located to lower pump and probe frequencies in the T-2D-IR data, consistent with the shift to lower frequencies observed for the CO stretching modes of **1** upon photolysis to form a pentacarbonyl species. This is attributable to the increased electron density back-donation to the remaining CO ligands. The broadening of the lineshapes caused by the cyanoheptane solvent leads to reduced resolution in the T-2D-IR data however and assignment of individual peak pairs is non-trivial. In order to accentuate the differences between the peaks due to the photoproduct and the parent molecule 2D bleaches, the 2D-IR spectral response (Fig 2(a)) was scaled and subtracted from the T-2D-IR data in Fig 2(b) to yield a double-difference spectrum (Fig 2(c)). The double difference spectrum thus contains only peaks due to the photoproducts, assuming a complete subtraction of the parent molecule peaks. In order to guide the eye, horizontal and vertical dashed lines from the 2D-IR and T-2D-IR spectra indicate parent molecule vibrational mode frequencies in Fig 2 so that the intersections of these lines on the double difference plot (Fig 2(c)) identify all of the parent molecule peak positions. Peaks not coinciding with these dashed guides are therefore assignable to photoproduct transitions.

The clearest region of the double difference spectrum where diagonal and off-diagonal photoproduct peaks are observed is highlighted by the red-dashed box. This corresponds to a vertical slice through the data at a probe frequency of 2050 cm^{-1} . This slice has been extracted and is shown in Fig 3. From fitting this slice to Gaussian lineshape functions, it is apparent that the diagonal peak is present at $\sim 2045\text{ cm}^{-1}$ alongside a broad band centred at 1979 cm^{-1} and a third, smaller feature located at 1930 cm^{-1} . These indicate that the spectral response of the photoproduct peaks shows a diagonal peak near 2050 cm^{-1} which is vibrationally coupled to modes near 1979 and 1930 cm^{-1} . In the case of the 1979 cm^{-1} transition, the increased width in comparison to the other two is suggestive of more than one overlapping transition as may be expected for a pentacarbonyl molecule lacking significant symmetry. It is noted that the combined effects of line broadening due to the solvent and the effects of convolution of the spectral lineshape with the pump pulse bandwidth profile significantly reduce the spectral resolution.

In the case of similar data recorded with heptane solution, photoproduct peaks were observed at 2050 , 1999 , 1978 and 1941 cm^{-1} .⁹ Assuming that the broad peak is in fact two overlapping resonances and allowing for a solvatochromic shift attributable both the polar solvent and a more strongly coordinating species it is reasonable to assign the above set of transitions to a photoproduct species. Comparison of these peaks with those observed in Fig 1(b) therefore indicate that the transitions observed at 2056 and 2008 cm^{-1} are likely to be due to vibrational cooling as they show no coupling to the other photoproduct transitions. This is also consistent with their positions slightly to the red of the main photoproduct bleaches, implying that they belong to hot ground state species.

Conclusions

T-2D-IR spectroscopy has been employed to determine the infrared absorption frequencies and coupling patterns of the photoproduct vibrational modes of the species formed following photolysis of **1** in cyanoheptane. Double difference T-2D-IR analysis has revealed a number of peaks due to the photoproduct and they show spectroscopy consistent with the formation of a pentacarbonyl solvent adduct species. In the data presented here line broadening effects limit the ability to resolve all photoproduct diagonal and off-diagonal peaks, however the similarities between this data and that obtained in heptane are sufficient for a conclusion to be reached. Future

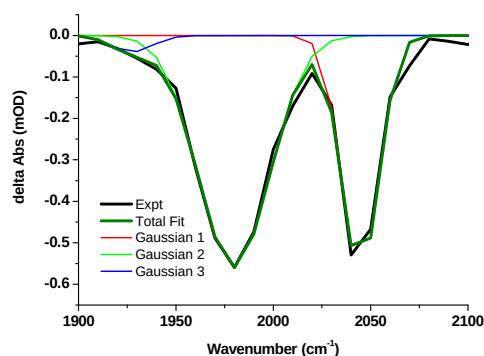


Figure 3: Slice through the T-2D-IR double difference spectrum in Fig 2(c) at a probe frequency of 2050 cm^{-1} . Thin lines show the results of fitting to three Gaussian lineshape functions.

studies using photon echo-derived 2D-IR methods under development will provide the improved spectral resolution required for observation of all the individual 2D peaks.

Acknowledgements

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Ultrafast TRIR Studies on Tetraazidosilicon Complexes

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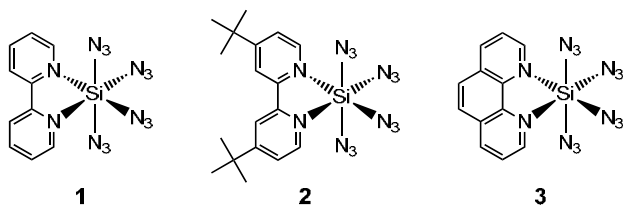
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Introduction

Finding compounds that can both reliably store and release energy in a controlled manner is a great challenge. Nitrogen-rich compounds are often highly endothermic and, as high-energy density materials (HEDMs), store a lot of chemical energy.¹⁻³ Nitrogen-rich HEDMs have several superior properties over conventional explosives, propellants and pyrotechnics, mainly due to the fact that they release less smoke, toxic byproducts and green house gases.⁴ Their main product of decomposition is dinitrogen. There are several ways to trigger energy release – reactions with light being one of them. Nitrogen-rich molecules, however, constitute a photochemically not well-investigated area.

Recently, we have prepared hexacoordinate tetraazido silicon complexes ³ (**1-3**, Scheme 1) as a new type of nitrogen-rich compounds. These complexes contain diimine ligands as chromophores and azido ligands. They are less friction sensitive, moderately air sensitive, colourless solids, which are soluble in a number of aprotic solvents. The azido ligands act as reporter groups and make these compounds ideal for mechanistic photochemical studies using time-resolved infrared (TRIR) spectroscopy.



Scheme 1.

Preliminary results of picosecond (ps) and nanosecond (ns) TRIR experiments with solutions of $\text{Si}(\text{N}_3)_4(\text{bpy})$ (**1**), $\text{Si}(\text{N}_3)_4(\text{tBu}_2\text{bpy})$ (**2**) and $\text{Si}(\text{N}_3)_4(\text{phen})$ (**3**) are presented here as part of our ongoing EPSRC funded research program *Real-time structural dynamics of molecular systems for energy generation and storage*.⁵

Experimental arrangements

The complexes **1-3** were synthesized in adaptation of published procedures.³ The PIRATE⁶ and ULTRA⁷ spectrometers were used for the ps- and ns-TRIR measurements. Solutions were prepared in dry degassed solvents, saturated with nitrogen and kept under slight pressure in a recirculating flow system at r.t. The optical density in the photolysis cells at $\lambda_{\text{exc}} = 266 \text{ nm}$ was kept below unity.

TRIR spectroscopy

TRIR spectra of the complexes **1-3** were obtained in acetonitrile (MeCN), tetrahydrofuran (THF) and CH_2Cl_2 solutions under 266 nm excitation, and registered in the region between 1920 cm^{-1} and 2260 cm^{-1} . In the following, the temporal evolution of the TRIR spectra of complex **1** (Scheme 1) is described in more detail.

The IR spectra of the largely C_2 symmetric tetraazido complexes **1-3** feature generally three intense bands in the mid infrared region which arise from four ‘asymmetric’ azide N-N-N vibrations of the symmetry species A and B. In THF solutions of complex **1** the parent bands at 2212, 2125 (shoulder) and 2149 cm^{-1} are fully bleached 4 ps after laser flash (266 nm) and several broad transient bands are present. These transients are centred at 1992 cm^{-1} (species X) and overlapped with the parent bleaches with apparent maxima at 2089 and 2138 cm^{-1} (Y) and 2161 cm^{-1} (Z) (Fig. 1, left).

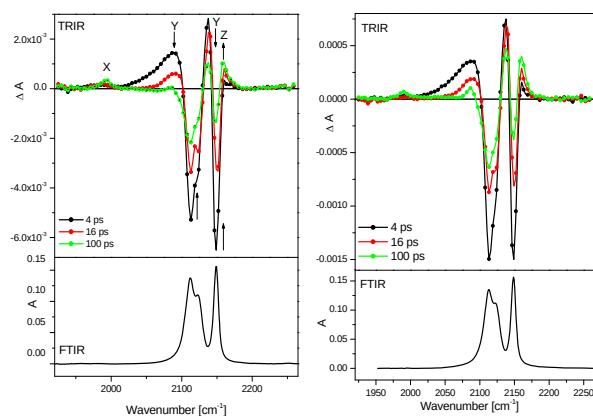


Figure 1. TRIR spectra in THF solution ($\lambda = 266 \text{ nm}$, top row) at $t = 4, 16$ and 100 ps of $\text{Si}(\text{N}_3)_4(\text{bpy})$ (**1**, left) and $\text{Si}(\text{N}_3)_4(\text{phen})$ (**3**, right) and respective ground state FTIR spectra (bottom row).

Over the next 100 ps the bands associated with the transient Y (at 2089 and 2138 cm^{-1}) narrow and decay very fast, with the parent bleach bands recovering synchronously. The band narrowing and a hypsochromic of the single band of species X can also be discerned. A kinetic analysis of the decay of Y and the recovery of the parent estimated the lifetime for this process to values between 8 and 18 ps. Furthermore, the rise of an initially small transient band at 2162 cm^{-1} (Z) occurs during the decay of the transient Y (Fig. 2, red trace).

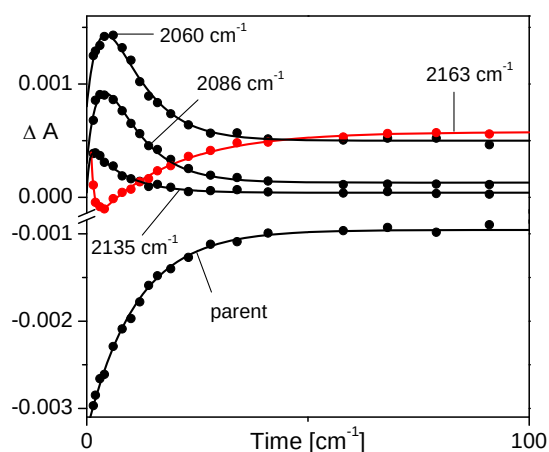


Figure 2. Kinetic traces of **1** in THF solution between 1 ps and 100 ps showing the decay of transients Y (black), the rise of transient Z (red) and partial recovery of a parent band.

In order to obtain more accurate spectral information the parent bleach bands were subtracted from the ps-TRIR spectra and the residual fitted using Voigt profiles (Fig. 3, left). For the TRIR spectrum at 100 ps this treatment afforded accurate peak maxima and approximate line widths for four transient bands at 1992 cm^{-1} (X), 2106, 2133 and 2156 cm^{-1} (Z). The bands associated with transient species Z persist throughout the investigated time delays up to 5000 ns. The single band of X can be assigned easily to the uncoordinated N_3^- ligand or azide anion N_3^- , by comparison with FTIR spectra of THF solutions of salts containing this anion (e.g. $\text{N}(\text{PPh}_3)_2^+ \text{N}_3^-$, 1993 cm^{-1}). The remaining bands (Z) arise from species with coordinated azido groups. See Table 1 for an overview of band positions.

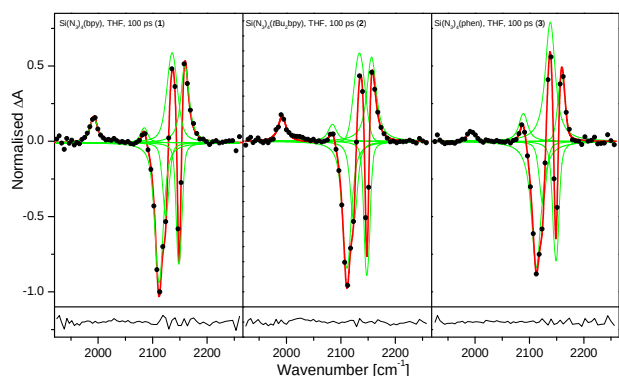


Figure 3. Difference TRIR spectra in THF solution ($\lambda = 266$ nm, top row) at $t = 100$ ps of compounds **1** (left), **2** (middle) and **3** (right). Spectra are normalised to the total area of the bleached parent absorptions; spectral points (black), Voigt profiles (green), fit curve (red), fit residues (bottom row).

The potential presence of anionic products prompted an investigation of the effect of polarity and coordination ability of the solvent. Studies performed in THF and CH_3CN revealed only minor differences in the position and temporal evolution of the transient bands observed in solutions of **1** in THF and MeCN on the 1 ps to 100 ps time scale (Fig. 4, Table 1). However, the ratio of band areas (N_3^- : transient) changes significantly from 1 : 7 in THF to 1 : 3 in MeCN. Hence, the stronger coordinating and more polar solvent (MeCN) promotes the formation of ionic photoproducts. The diimine ligand too has a marked, though less well understood, influence on the ratio of photoproducts. The 100-ps TRIR spectra in THF show a

significantly enhanced formation of N_3^- for (4,4' disubstituted) bpy over phen complexes as shown by the ratios of band areas, 1 : 7 (bpy), 1 : 7 ($t\text{Bu}_2\text{bpy}$), 1 : 27 (phen).

In THF the N_3^- transient band (X) vanishes with a lifetime of 633 ± 55 ns which is matched by the synchronous recovery of the parent bands. Parent recovery during this period amounts to only 40% of the total bleach and a close investigation of the features of the transients at 2106, 2133 and 2156 cm^{-1} reveals more subtle changes. These changes could be accounted for by expanding the model of Voigt profiles by two additional, less intense transients (Z') (see Table 1), which in the TRIR spectrum remain hidden throughout the observed times delays. A global curve fit using the expanded model reveals that the bands of Z remain unchanged while those of Z' decay completely. A kinetic analysis of the associated band area changes reveals that both parent recovery and the decay of the bands of X and Z' are synchronous on this time scale (Fig. 5).

A tentative mechanism can be drawn up at this stage (Scheme 2). Upon excitation of complex **1** an excited state is formed (transient Y), which fully decays within the first 50 ps. The decay leads in part to the reformation of the parent complex. The remainder is involved in photodissociation and reacts upon breaking a Si- N_α bond and formation of the azide anion (transient X) and a corresponding cationic silicon complex (transient Z') (i), and also upon breaking a $\text{N}_\alpha\text{-N}_\beta$ bond within one azido ligand generating dinitrogen gas and a nitrene complex (transient Z) (ii). N_α denotes the ligating N atom of the azido ligand. Recombination of the ionic products makes process (i) fully reversible. Reattachment of dinitrogen, however, is thermodynamically strongly disfavoured and hence process (ii) is irreversible. The presence of three long-lived transient bands (Z) points to a complex bearing three azido ligands. Preliminary results of DFT calculations (Dr. A. Meijer) indicate that a nitrene complex (as postulated in Scheme 2) is not a minimum on the energy hypersurface. Once the $\text{N}_\alpha\text{-N}_\beta$ bond is broken, the resultant 'nitrene' inserts into an adjacent Si-N bond.

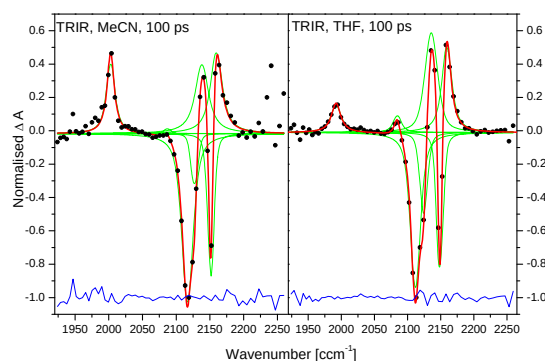


Figure 4. Difference TRIR spectra in THF solution ($\lambda = 266$ nm) at $t = 100$ ps of **1** ($\text{Si}(\text{N}_3)_4(\text{bpy})$) in MeCN (left) and THF (right). Spectral points (black), Voigt profiles (green), fit curve (red), fit residues (blue lines).

Inspection of the combined ns- and ps-TRIR spectra of the related complexes **2** and **3** suggests that analogous mechanisms operate in the photoreactions of the latter.

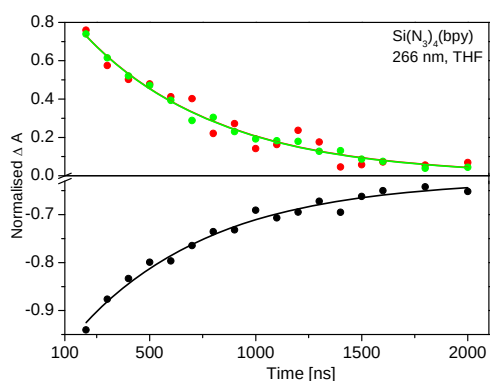
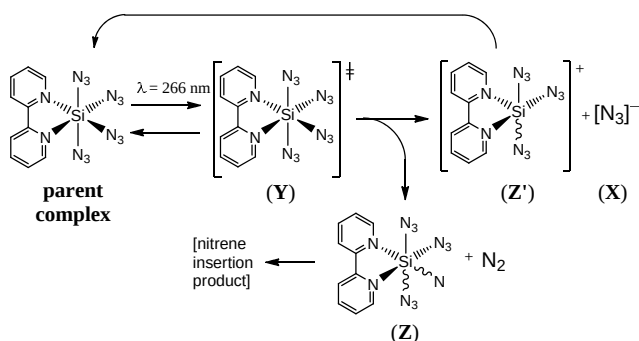


Figure 5. Kinetic traces of normalized band areas in a THF solution of from 200 ns to 2000 ns showing the synchronous decay of the transient bands of X (green) and Z' (red) and the recovery of the parent complex (black).



Scheme 2. Proposed reaction mechanism.

Conclusions

The photochemical behaviour of several new tetrazido silicon complexes has been investigated with the aim to identify a mechanism for photochemical energy release and the generation of dinitrogen. The precise nature of several transients is still unknown. Using the transient absorption capabilities of the ultrafast laboratory at the CLF we plan to further investigate the possibility of an electronic excited state being formed initially prior to breaking either Si-N α or N α -N β bonds. Quantum chemical calculation performed in Sheffield (Dr. A. Meijer) on the minimum structures of the candidate species (Scheme 2) will provide frequencies for the asymmetric N-N-N stretch vibrations, which will then be used to match with the bands found in the TRIR experiments. The presented study has shown that using ps and ns TRIR spectroscopy is suited to unravel the mechanism of photochemically induced reaction in nitrogen-rich compounds.

Table 1. Band position [cm⁻¹] of ground states and transients determined from time resolved infrared spectra at ca. 30 ps (λ_{exc} = 400 nm) using pseudo Voigt profiles.

Si(N ₃) ₄ L L =	Parent Bands	Transient Bands
Solvent: CH ₃ CN		
bpy	2116, 2127, 2151	2002 (X) 2087, 2138, 2159 (Z)
^t Bu ₂ bpy	2115, 2125, 2150	
phen	2116, 2126, 2150	2005 (X) 2092, 2135, 2158 (Z)
Solvent: CH ₂ Cl ₂		
bpy	2115, 2127, 2152	-
phen	2116, 2127, 2152	-
Solvent: THF		
bpy	2212, 2125, 2149	1992 (X) 2106, 2133, 2156 (Z) (2121, 2156) (Z')
^t Bu ₂ bpy	2111, 2124, 2148	
phen	2113, 2125, 2149	1993 (X) 2089, 2139, 2161 (Z) (2127, 2158) (Z')

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Introduction

Organometallic complexes have found many applications in organic syntheses. For instance, coordination of an arene unit to a $M(CO)_3$ ($M = Cr$ or Mo) fragment can activate the arene toward a variety of reactions.¹ Cobalt complexes, such as $(\mu_2\text{-alkyne})Co_2(CO)_6$ have been widely used in the Pauson-Khand reaction.² The PKR is a versatile route to the synthesis of natural products and pharmaceutically active compounds.³ We have recently demonstrated that the Pauson-Khand can be driven using visible light irradiation at ambient temperatures.⁴

$(\eta^6\text{-Benzene})Cr(CO)_3$

The photochemistry of $(\eta^6\text{-Benzene})Cr(CO)_3$ has been investigated since the early days of flash photolysis^{5,6} and matrix isolation.⁷ Our recent TRIR studies in room temperature *n*-heptane identified a vibrationally cold excited state as the precursor to CO expulsion to form $(\eta^6\text{-Benzene})Cr(CO)_2$.⁸ This process is comparatively slow, taking place over the course of 150 ps ($\lambda_{exc.} = 400$ nm).

Within 1 ps of excitation pulse the two ν_{CO} bands of the parent $(\eta^6\text{-Benzene})Cr(CO)_3$ complex at 1984 and 1916 cm^{-1} were depleted and two broad features formed, at ~ 1965 and 1887 cm^{-1} assigned to the excited state species, $[(\eta^6\text{-Benzene})Cr(CO)_3]^*$. This species is formed with excess vibrational energy. Within 3 ps the bands become narrow and the ν_{CO} bands of “cold” $[(\eta^6\text{-Benzene})Cr(CO)_3]^*$ appear at 1965 and 1887 cm^{-1} . The rate of this process was found to be $1.6 (\pm 0.3) \times 10^{12} s^{-1}$ at 298 K.

The effect of temperature change on this process (274 to 302 K) yielded an activation enthalpy ($\Delta H^\ddagger$) of $-10 (\pm 4) kJ mol^{-1}$, suggesting a barrierless process. The activation entropy ($\Delta S^\ddagger$) is $-50 (\pm 16) J mol^{-1} K^{-1}$.

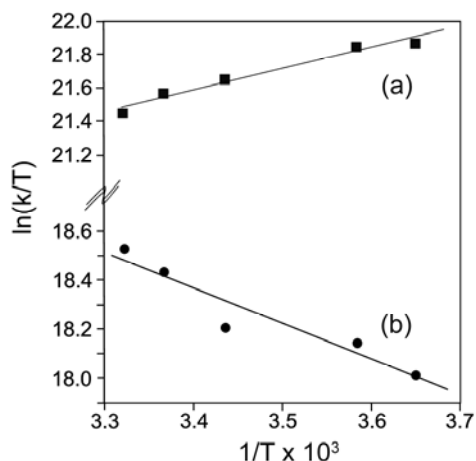


Figure 2. Eyring plots for (a) the growth of the absorption band at 1887 cm^{-1} i.e. the cold excited state and plot (b) was obtained from the growth of the 1872 cm^{-1} band of $(\eta^6\text{-Benzene})Cr(CO)_2$ (3).

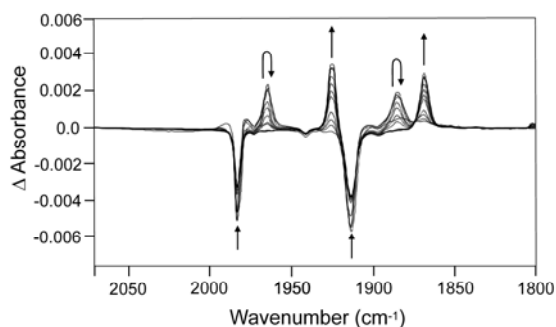
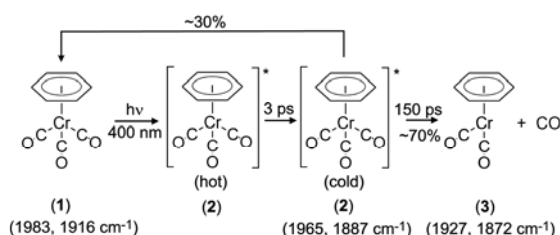


Figure 3. The TRIR spectra obtained following 400 nm excitation of $(\eta^6\text{-Benzene})Cr(CO)_3$ in *n*-heptane, spectra were recorded at 1, 7, 12, 21, 51, 70, 106, 160, 350, 500, 750, 1000 ps after the excitation pulse. Arrows indicate the time-dependent behavior of the absorption features.

During the subsequent 150 ps, $[(\eta^6\text{-Benzene})Cr(CO)_3]^*$ decays with the concomitant formation of $(\eta^6\text{-Benzene})Cr(CO)_2$ and reformation of $(\eta^6\text{-Benzene})Cr(CO)_3$. $(\eta^6\text{-Benzene})Cr(CO)_2$ has ν_{CO} bands at 1927 and 1872 cm^{-1} . It appears that formation of the CO-loss species occurs mainly from the “cold” excited state as does regeneration of $(\eta^6\text{-Benzene})Cr(CO)_3$ (Scheme 1).

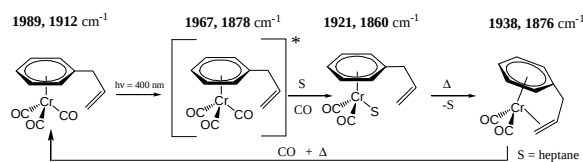
Eyring plots for the formation of $(\eta^6\text{-Benzene})Cr(CO)_2$ gave $\Delta H^\ddagger$ and $\Delta S^\ddagger$ values of $+12 (\pm 4) kJ mol^{-1}$ and $-4 (\pm 7) J mol^{-1} K^{-1}$ respectively.



Scheme 1 The excitation of $(\eta^6\text{-Benzene})Cr(CO)_3$ produces a vibrationally hot excited state. This cools over 3 ps to a vibrationally relaxed excited state which loses CO and relaxes to $(\eta^6\text{-Benzene})Cr(CO)_3$ over 150 ps.

$(\eta^6\text{-Allylbenzene})Cr(CO)_3$

$(\eta^6\text{-arene})Cr(CO)_3$ and $(\text{cyclopentadienyl})Mn(CO)_3$ are of interest in the design of ultrafast photoswitches.⁹ When the arene ligand contains a substituent with a functional group capable of coordinating to an electron deficient 16 electron CO-loss species, the CO-loss species undergoes a subsequent reaction with the pendant substituent to form a “chelate” species (Scheme 2).



Scheme 2

The TRIR data obtained following photolysis of $(\eta^6\text{-allylbenzene})\text{Cr}(\text{CO})_3$ are presented in Figure 4. Two excited state species are generated. The species at 1967 and 1878 cm^{-1} decays ($1.47 \times 10^{-12} \text{ s}^{-1}$) to generate the CO loss species (D). This subsequently forms the chelate species (C).

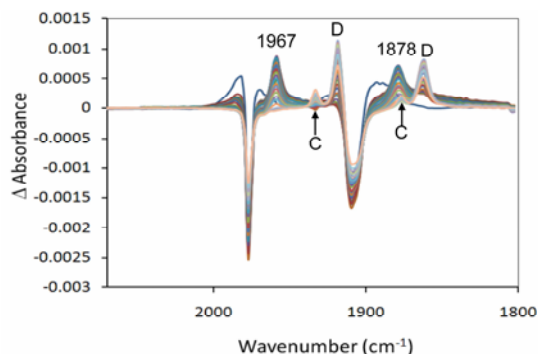


Figure 4 The TRIR spectra obtained following 400 nm excitation of $(\eta^6\text{-allylbenzene})\text{Cr}(\text{CO})_3$ in *n*-heptane.

$(\mu_2\text{-Alkyne})\text{Co}_2(\text{CO})_6$ complexes

The last section of this report deals with the photochemistry of acetylene cobalt carbonyl compounds. Irradiation of diphenylacetylene $\text{Co}_2(\text{CO})_6$ at 400 nm in THF resulted in depletion of the parent bands and the formation of bands at 2074, 2041 and 1992 cm^{-1} , which underwent vibrational relaxation within 10 ps. The product species was assigned to the triplet diradical which rapidly reacts within 80 ps, to regenerate the parent complex. No evidence for CO loss was obtained for any of the complexes in Figure 5. The photochemistry of complexes 1-3 was independent of the solvent used (pentane, THF, acetonitrile, or DCM)

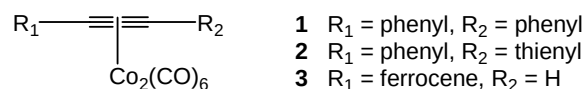


Figure 5. $(\mu_2\text{-alkyne})\text{Co}_2(\text{CO})_6$ complexes studied previously

The aim of this study was to increase the efficiency of the CO loss pathway, and to improve their use as CO-releasing molecules (CORMs). Some of the complexes investigated are shown in Figure 6.

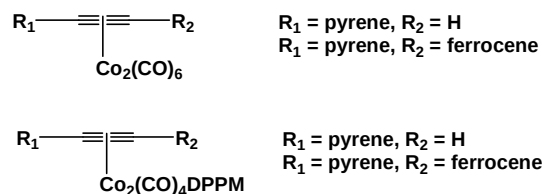


Figure 6. $(\mu_2\text{-alkyne})\text{Co}_2(\text{CO})_6$ complexes studied.

As previously all $(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_6$ and $(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_4(\text{DPPM})_2$ complexes gave rise to the triplet diradical species, $^3(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_6$ or $^3(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_4(\text{DPPM})_2$ with lifetimes between 26 and 78 ps. However the CO loss species, $(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_5(\text{solvent})$ was also observed following irradiation of both $(\mu_2\text{-pyrenylacetyleneferrocene})\text{Co}_2(\text{CO})_6$ and $(\mu_2\text{-pyrenylacetylene})\text{Co}_2(\text{CO})_6$ (Figure 7). The CO loss species for

the latter has ν_{CO} bands at 2074, 2030, 2007, and 1975 cm^{-1} , consistent with literature values. Infrared bands for this CO-loss species remain on the nanosecond timescale, and the parent bands do not fully recover on this timescale.

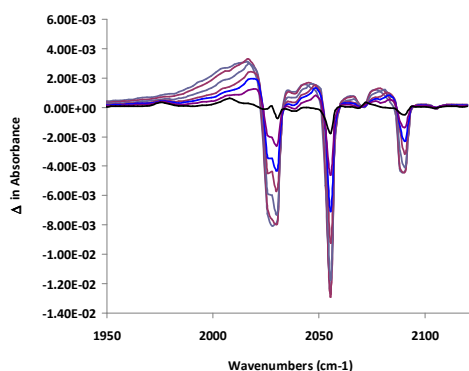


Figure 7. The TRIR spectra obtained following 400 nm excitation of $(\mu_2\text{-pyreneacetylene})\text{Co}_2(\text{CO})_6$ in *n*-heptane, at 5, 10, 15, 40, 60, 100 and 1000 ps.

Conclusions

The results presented here confirm that photo-induced CO-loss from $(\eta^6\text{-Benzene})\text{Cr}(\text{CO})_3$ occurs via a vibrationally relaxed excited state over a small enthalpy barrier caused by the avoided crossing of two excited state surfaces.

In the case of the $(\mu_2\text{-alkyne})\text{Co}_2(\text{CO})_6$ and $(\mu_2\text{-alkyne})\text{Co}_2(\text{CO})_4(\text{DPPM})_2$ complexes, both give rise to a triplet diradical species following irradiation. These react rapidly to regenerate the parent complex in < 80 ps. Photoinduced CO loss was observed following irradiation of both $(\mu_2\text{-2-pyrenylacetylene})\text{Co}_2(\text{CO})_6$ and $(\mu_2\text{-2-pyrenylacetyleneferrocene})\text{Co}_2(\text{CO})_6$ complexes.

Acknowledgements

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Excited-State Dynamics of Adenine Thymine Dinucleotides: Influence of Stacking

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Introduction

The DNA double helix contains the information required for replicating life; molecular interrogation of the structure through transcription and translation relating it to function. The integrity of this process depends on the stability of DNA and its ability to resist and/or repair erroneous modification. For this reason the study of the excited state dynamics of DNA is of great interest.^{1,2,3,4} Through our research we hope gain understanding of the influence of structure on the properties of long-lived excited states achieved through base stacking and hydrogen-bonding interactions and further understand the ultrafast processes that may lead to genetic mutation.

Picosecond ultrafast transient infrared (ps-TRIR) provides structural as well as kinetic information and has been used to study the photodynamics and the photoreactions of mononucleotide and polynucleotide systems.^{5,6} Until very recently the singlet excited states of nucleotides were thought to occur on a subpicosecond timescale. However, using ps-TRIR we identified a longer-lived transient state (34 ps) in 5'-dCMP, which had been inferred from transient absorption work by Hare et. al, and provisionally assigned it to the $^1n_N\pi^*$ non-emissive - dark state.^{7,8} Our ps-TRIR measurements of the other nucleotides revealed solely information on vibrational cooling.⁹ However, polynucleic acids display longer-lived excited states and there is debate regarding the origin of long-lived excited in states and the role of stacking interactions. In this report we present further findings from our psTRIR study of excited state dynamics of the two A-T dinucleotides, namely 2'-deoxyadenyl-(3'-5')thymidine (dApT), and thymidyl-(3'-5')-2'-deoxyadenosine (Tp dA), see Figure 1. These structural isomers exhibit different stacking behaviour in solution and as such were chosen to investigate the role of stacking on the excited state where the nucleic acid components remain constant.

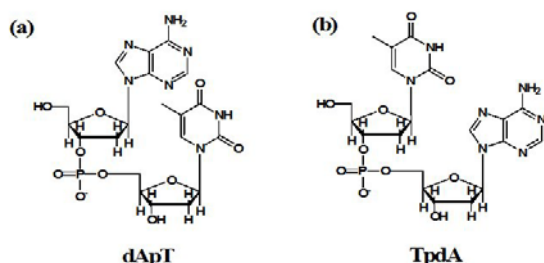


Figure 1: The structure of the two dinucleotide systems studied, (a) dApT and (b) Tp dA.

Experimental

Measurements were made using the picosecond infrared absorption and transient excitation (PIRATE) system.¹⁰ The data were collected in a number of 150 cm^{-1} spectral windows

using a delay line for optical delays between 2 ps and 1 ns. The difference signal was calibrated using water vapour lines present in the probe spectrum, and the spectral windows were interleaved using overlapping transients recorded at the same delay time. The sample was raster scanned in the x- and y-directions at an approximate rate of 100 mm s^{-1} . The 1-pixel accuracy was $\pm 4 \text{ cm}^{-1}$. Ground state UV and FTIR measurements were recorded using a Perkin-Elmer Lambda2 and a Nicolet Avatar 360 respectively. Global analysis of the relaxation behaviour was achieved by simultaneous fitting of the transient absorption decay curves at different wavelength using well-known global fitting procedures¹¹ and assuming a biexponential decay model. Briefly, global fitting with parameter sharing is an option of fitting multiple datasets simultaneously for one function (in this case biexponential decay model). For each shared (global) parameter (in this case lifetime), the one best-fit value that is estimated from all the fitted datasets is obtained.

Results and Discussion

Heterodinucleotide systems: dApT and Tp dA

At room temperature the dinucleotides exist in both stacked and unstacked forms in solution (dApT 38% and Tp dA 20%).¹² The infra red spectra for both dinucleotides shows bands characteristic of the adenine (ring vibrations ca. 1626 cm^{-1}) and thymine (ring and carbonyl stretches 1629, 1660 and 1684 cm^{-1}), see Figure 2. The ps-TRIR spectrum of dApT following UV excitation is shown in Figure 2a with the shorter delay times (2-10 ps) shown in red and the longer delays (15-1500 ps) shown in black. The spectrum shows bleaches centred at 1623 cm^{-1} , 1661 cm^{-1} and 1694 cm^{-1} and a broad transient spanning from 1530-1612 cm^{-1} . An important feature is the 'tracking' (i.e. shifting of the band to higher wavenumbers) of the transient bands at early times (red delays) and the persistence of bleached and transient bands after 15 ps (black delays) show the presence of a longer-lived species. This is in contrast to what is observed for the constituent mononucleotides, which undergo rapid ground state recovery over $2.2 \pm 0.2 \text{ ps}$ (5'-TMP) and $4.1 \pm 0.4 \text{ ps}$ (5'-dAMP). The relative intensity of the individual bands within the bleach region was found to change with time, indicating the presence of more than one species. At early time delays the ratio of the main bleach (1620 cm^{-1}) to the thymine-based bleach (1661 cm^{-1}) is 2:1 but at later times the ratio of the bleaches becomes 1:1 and remains so as the sample relaxes back to the ground state. The kinetic analysis of dApT was carried out using standard global fit analysis of the transients and bleach bands (see experimental section). This showed the presence of two species with lifetimes of $3.9 \pm 0.4 \text{ ps}$ and $75 \pm 7 \text{ ps}$. The first lifetime is characteristic of the monomer species and therefore attributed to the unstacked dinucleotide, while the second lifetime component is attributed to the stacked form of the dinucleotide in solution. The broad absorption band for this longer lived species, which is presumed to be due to an

excited state dimer, is similar to that observed in the polynucleotide {polydA-dT}₂.¹²

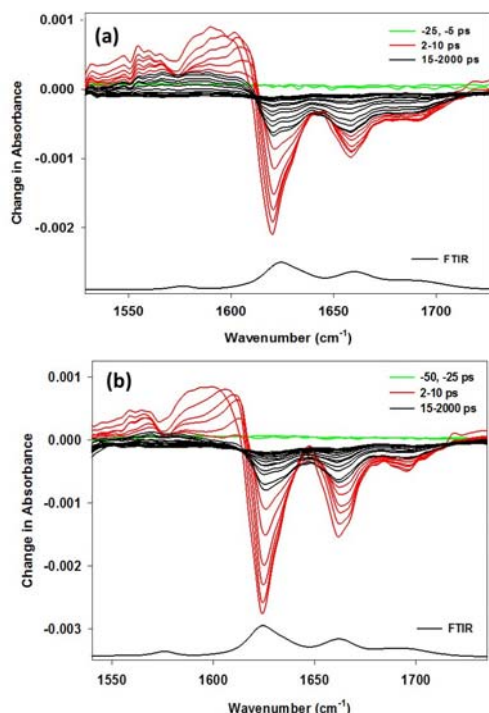


Figure 2: ps-TRIR spectra following UV excitation (300 fs, 267 nm) of 10 mM nucleotide systems in 50 mM potassium phosphate D₂O buffer at pH 7, at 20° C. (a) dApT and (b) TpdA. Delays are at -50, -25 (green), 2-10 (red), 15- 2000 ps (black). Below each data set is the FTIR groundstate spectrum taken prior to the ps-TRIR experiment.

The ps-TRIR spectrum of TpdA following UV excitation is shown in Figure 2b. As for dApT the spectrum shows bleaches centered at 1623 cm⁻¹, 1661 and 1694 cm⁻¹ and a broad transient spanning from 1530-1612 cm⁻¹ are recorded. Again the persistence of the bleached and transient bands after 15 ps indicated the presence of a long-lived species. Again kinetic analysis yielded two lifetimes for recovery of 4.2 ± 0.4 ps and 50 ± 5 ps. The former value is in close agreement to that found for the short-lived species in dApT (3.9 ± 0.4 ps) as would be expected if they arise from the unstacked nucleotides. However, the long-lived species shows a significantly shorter lifetime compared to that of its isomer (75 ± 7 ps). Furthermore, the relative yield of the long-lived transient was determined to be approximately 50 % of that found for dApT, consistent with the smaller amount of stacking in the TpdA isomer. The long-lived species possessed a distinctive spectroscopic signature in the region 1500-1600 cm⁻¹, which is not present in the homodinucleotides, dApdA and TpT. The signature is identical to that of the long-lived state observed in double stranded poly(dA-dT). Indicating that the excitation in the polymer is localized on the dinucleotide unit.

Conclusions

This study illustrates how ps-TRIR can provide valuable insights into the fast photophysical properties of nucleic acids, and so complement the measurements obtained by the more established transient Uv/visible absorption and luminescence methods.⁵ In particular it is apparent from this study that stacking of the nucleobases gives rise to a longer-lived species, which possesses a characteristic infra-red band in the 1550 – 1600 cm⁻¹ region in the case of the adenine-thymine bases. (The yield of the long-lived transient is greater for dApT than for TpdA reflecting the larger amount of stacked form present at

equilibrium in solution.) Takaya et al.¹³ recently made femtosecond Uv-vis transient absorption measurements on dinucleotides and assigned long-lived states to purine-pyrimidine charge transfer states. The characteristic signal we observe may therefore be associated with such a charge redistribution in the stacked bases. In this connection we may point out we have also observed that long-lived excimer excited states from adenine homodinucleotides do not show a similar infra-red absorption. Such differences in the spectroscopic signals opens up the prospect of being able to identify localised states in photo-excited DNA – for example, we have previously suggested that other localised states may be identifiable in {polydG-dC}₂.¹⁴

The kinetic measurements firstly reveal the major difference in properties between the stacked and unstacked forms of the dinucleotide, with the unstacked conformers behaving very much like the mononucleotides, which undergo sub-picosecond relaxation to the ground states. The ‘hot’ ground state formed is observed to deactivate with a lifetime of about 4 ps and as expected the rate of vibrational deactivation does not depend on the isomeric species studied. However there is a difference in the rate of relaxation for the longer-lived excited states of the two AT isomers and these differences in lifetimes appear to correlate with the extent of stacking.

In conclusion this study shows that a study of dinucleotides can be very useful in understanding the role of stacking of the nucleobases, in the absence of H-bonding base-pairing, and as such should be helpful in allowing us to determine the relative significance of intra- and inter-chain processes in double-stranded DNA.¹⁵

Acknowledgements

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 15. Aspects of this work were presented at the Ultrafast Phenomena XVI conference.

Antigens laid bare: the intrinsic conformation of Lewis^x

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Introduction

Lewis^x (Le^x), one of the so-called Type 2 antigens (Le^x, Le^y) presenting the trisaccharide epitope, Gal β 1-4[Fuc α 1-3]GlcNAc, is expressed by many cancers of epithelial origin and is an important immunotherapeutic target. Knowing its structural preference(s), particularly in its bioactive conformation, is a key requirement in designing drugs which might inhibit its binding and activity. Condensed phase environments are known to favour relatively rigid, compact structures with the terminal pyranosides, Gal and Fuc, stacked in parallel and separated – depending on the environment – by approximately 4 to 5 Å. Thus structures determined through X-ray diffraction can be stabilised by the protein or crystalline environment and NMR measurements of nuclear Overhauser effects, scalar or residual dipolar couplings in aqueous environments, coupled with MD simulations to provide dynamically averaged structures, can be influenced by explicit hydration and ‘water bridging’ across the glycosidic linkages. To understand the influence of the environment on the structure of Lewis antigens it is essential to know first, their *intrinsic* conformational landscapes but these have not been determined experimentally. MM and MD calculations have been conducted ‘*in vacuo*’ but their predictions depend on the accuracy of the assumed force fields; *ab initio* calculations of Le^x have also been conducted, but only at a low (Hartree-Fock) level of theory.

The vibrational spectra of individual carbohydrate conformers can be obtained in a cold molecular beam using double resonance IR-UV ion dip spectroscopy (IRID),^[1] and by comparing them with those determined through a combination of molecular mechanics, density functional theory (DFT) and *ab initio* calculations it is possible to determine the intrinsic carbohydrate structures experimentally. Following this strategy, the target molecule, Le^x has been synthesized in large scale, as the β -O-phenyl glycoside (to provide the necessary UV chromophore required for IRID detection, see Figure 1).

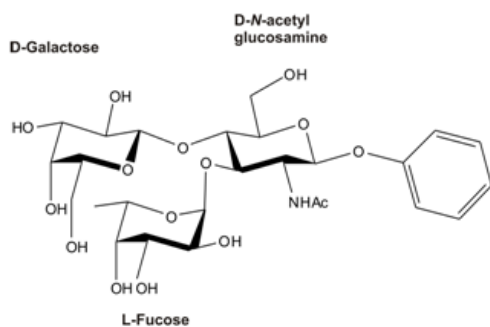


Figure 1. Schematic molecular structure of the antigen, Lewis^x ('tagged' with a phenyl group to facilitate its spectroscopic interrogation).

Results and Discussion.

The computed structures and relative energies of the three lowest lying conformers are presented in Figure 2. The conformer that most closely resembles the ‘stacked’ structures (of the β -O-methyl glycoside) in aqueous solution, in the hydrated crystal and in protein-bound Le^x, does not correspond to the intrinsic global minimum predicted by the calculations but to the conformer calculated to lie at an energy 2.9 kJ mol⁻¹ above it, see Figure 3.

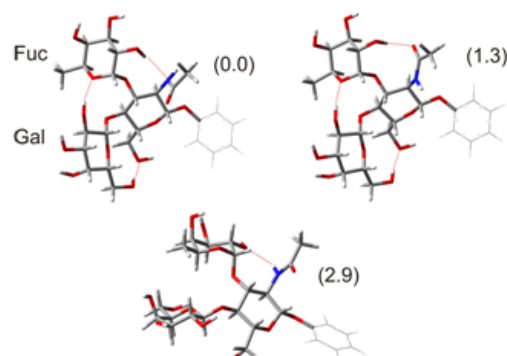


Figure 2. Structures and relative energies, in kJ mol⁻¹, of the three lowest energy conformers of O-phenyl Le^x.

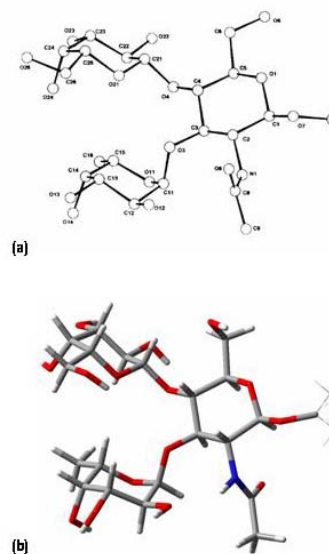


Figure 3. Comparison between (a) the crystal structure of β -O-methyl Lewis^x [2] and (b) the computed structure of the ‘stacked’ conformer, #3, of β -O-phenyl Lewis^x

Neither of the two lowest energy conformers displays this type of structure. In the stacked structure, apart from a weak hydrogen-bond between the GlcNAc and Fuc residues, $r(\text{NH}\cdots\text{OH}_2)=2.18\text{\AA}$, there are no inter-ring hydrogen bonds. This contrasts sharply with the other two structures, both of which display strong inter-ring linkages between all three residues, including: $\text{OH}_2^{\text{Gal}}\rightarrow\text{O}_5^{\text{Fuc}}$, $r(\text{OH}_2\cdots\text{O}_5)=1.83\text{--}1.88\text{\AA}$ and $\text{OH}_6^{\text{Gal}}\rightarrow\text{O}_6^{\text{GlcNAc}}$, $r(\text{OH}_6\cdots\text{O}_6)=1.99\text{\AA}$. These inter-ring linkages create quite rigid and inflexible structures but the glycosidic structure of the stacked conformer should be rather more mobile. The acetamido group is oriented roughly perpendicular to the ‘plane’ of the pyranoside ring (dihedral angle, C3-C2-N-H $\sim 85^\circ$) but the exocyclic CH_2OH groups adopt a ‘parallel’ *gt* orientation in almost every case apart from the stacked conformer, where there is no inter-ring hydrogen bonding and the orientation of the $\text{CH}_2\text{OH}^{\text{GlcNAc}}$ group is *gg*. Are these predictions in agreement with experiment?

The UV R2PI spectrum of the trisaccharide, vaporized through pulsed laser ablation and entrained in a cold Ar molecular beam, presents a broad, structureless bimodal band centred at $\sim 272\text{ nm}$ but the associated IRID vibrational spectra recorded with the UV probe laser located at different wavelengths within the band contour are all identical, suggesting the population of a single conformer only (unless they have identical UV absorption spectra). This suggestion is reinforced by an analysis of the experimental IRID spectrum, recorded between 3300 cm^{-1} and 3700 cm^{-1} and shown in Figure 4, together with the vibrational spectra predicted for each of the three lowest lying conformers. The experimental spectrum in no way resembles that of the stacked configuration, relative energy 2.9 kJ mol^{-1} , shown in Figure 3 - or those lying at higher relative energies, and it can be firmly excluded. It does resemble, but is in rather poor correspondence with the spectrum of the second conformer, relative energy 1.3 kJ mol^{-1} and the spectrum in closest correspondence with experiment and the only one to predict the closely spaced doublet at $3500\text{--}3550\text{ cm}^{-1}$, is associated with the global minimum conformation. The cluster of OH bands located at high wavenumbers, between $\sim 3570\text{ cm}^{-1}$ and 3650 cm^{-1} can be assigned to the weakly hydrogen-bonded peripheral ‘spectator’ OH groups. The more intense red-shifted doublet between 3500 cm^{-1} and 3550 cm^{-1} , indicates two moderately hydrogen bonded OH stretch modes, which can be assigned to $\sigma_2(\text{Fuc})$ and $\sigma_6(\text{Gal})$. The sharp feature at 3455 cm^{-1} is associated with the ‘free’ NH vibration and the strongly displaced and broadened band centred $\sim 3430\text{ cm}^{-1}$, which indicates a strongly hydrogen bonded OH group, can be assigned to the vibrational mode, $\sigma\text{OH}_2(\text{Gal})$.

These results confirm the theoretical predictions and reveal a clear distinction between the *intrinsic* conformational preference of Le^x , ‘*in vacuo*’ and the preferences established in aqueous, hydrated crystalline or protein-bound environments. They also present a conundrum. In the condensed phase, all the experimental evidence points to the adoption of a rather rigid, compact stacked conformation, despite the absence of intramolecular hydrogen bonding across the glycosidic linkages. In the gas phase these linkages do provide rigidity but the intrinsic molecular conformation does not correspond to the stacked configuration. In aqueous solution and as a bound ligand, the $\text{CH}_2\text{OH}(\text{Gal})$ orientation is *gt*, but in the gas phase it is *gg*. Some involvement of the aqueous environment is evidently implicated. Stabilisation of the higher energy stacked configuration is unlikely to be promoted by the solvent polarity since its calculated dipole moment, 2.6 Debyes, is much smaller than that of the global minimum conformation populated in the gas phase, 7.2 Debyes. Instead the preference for the stacked conformation in aqueous environments and its rigidity must be associated with the replacement of intramolecular hydrogen bonding by explicit hydration or ‘bridging events’ – in agreement with recent predictions made for the Lewis antigens on the basis of MD simulations using the GLYCAM04 force

field [3] and more broadly for oligosaccharides in general. The new gas phase results reported here, for Le^x , also imply a key structural role for an aqueous environment, including perhaps hydrophobic and electrolytic interactions, in controlling the conformation of Le^x and in consequence, its biological function.

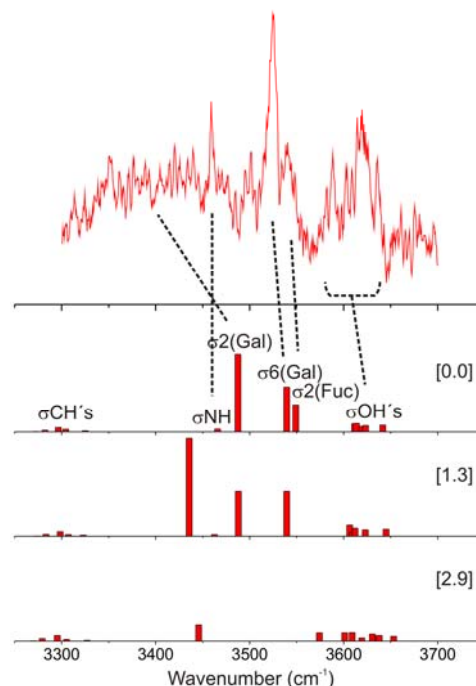


Figure 4. The experimental IRID spectrum of β -O-phenyl Le^x (upper trace) and the computed vibrational spectra of its three lowest energy conformers; relative energies (kJ mol^{-1}) shown in brackets.

Conclusions

The conformation of the Lewis^x antigen has been determined, free of environmental perturbation, at low temperature in the gas phase to reveal a global minimum energy structure lying below the rigid stacked conformations adopted in aqueous solution, in the crystalline hydrate or when bound to proteins. The result suggests that water plays a key structural role in controlling the conformation of Lewis^x and in consequence, its biological function.

Acknowledgements

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Carbohydrate molecular recognition: probing CH- π interactions

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Introduction

Molecular recognition of carbohydrates by proteins often involves their selective binding at sites adjacent to aromatic amino acid residues and X-ray crystallographic measurements have revealed a stacked, “apolar CH- π bonding” motif, with the aromatic side group providing a platform for the bound ligand. “Hydrophilic” OH- π hydrogen bonded interactions have also been inferred from the crystallographic data. IR ion dip (IRID) spectroscopy of O-H vibrational modes has been used in small model carbohydrate-aromatic systems isolated in the gas phase and free of neighbouring water molecules, to provide a more direct experimental probe of intermolecular bonding since their frequencies and intensities are extraordinarily sensitive to their local inter (and also intra) molecular hydrogen bonded environments. OH- π hydrogen bonding was signaled by a red-shift, broadening and intensification of one (or more) of the O-H bands.^[1] The *absence* of any significant displacement of the OH modes in an aromatic complex was taken as an indirect indication of carbohydrate binding through “CH- π ” interactions, provided the OH- π interaction was stronger than any pre-existing OH-O interaction with a neighbouring OH group; (the operation of CH- π interactions did not necessarily preclude a minor contribution from OH- π bonding as well).

Similar spectroscopic measurements of shifts in the IR frequencies of the C-H modes between free and bound carbohydrates, which might be used to ‘report’ directly on the incidence of CH- π bonding in their aromatic complexes, have not been undertaken. The observation of specific displacements might, with the aid of predictive quantum chemical calculations, help in their structural assignment since the magnitude and direction of the spectral shifts that can be associated with CH- π bonding have been intensively discussed in recent years, particularly the information they may convey about the nature of the interaction.

Two complexes which had previously been associated with CH- π , rather than OH- π bound structures because of the absence of any significant change in the OH spectrum of the bound carbohydrate, included methyl α -D-galactopyranoside•toluene (MeGal•Tol) and its 6-deoxy analogue, methyl α -L-fucopyranoside•toluene (MeFuc•Tol), see Figure 1; (the enantiomeric choices reflect Nature’s preferences). These experiments have now been extended using MeFuc•Tol as a benchmark system and toluene- d_8 , to obtain the IR spectrum of the bound carbohydrate in the CH stretch region without spectral interference from its aromatic partner. At the same time DFT calculations have been conducted for both the isolated and the complexed carbohydrate, using Zhao and Truhlar’s MO5-2X functional^[2] (which takes proper account of electron exchange and correlation) and the predictions have been compared with experiment, both for the OH spectra reported earlier and for the newly recorded CH spectra.

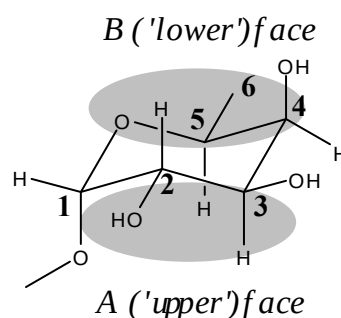


Figure 1. Schematic structure of methyl α -L-fucopyranoside (MeFuc)

Results and Discussion

The lowest lying structures (<10 kJ mol⁻¹ above the minimum) of MeFuc•Tol- d_8 calculated using the MO5-2X functional are listed, together with their relative energies and binding energies, in Table 1. ‘CH₁- π ’ or ‘OH₁- π ’ identify the character of the intermolecular interaction with the aromatic partner. Seven of the eight calculated structures are bound through CH- π interactions, leading to binding energies in the range $\sim(13 - 18)$ kJ mol⁻¹. The only one involving an OH- π hydrogen bond has the highest relative energy, 7.6 kJ mol⁻¹ and the rest involve binding either at the CH₁ site, which is located on the lower face of the pyranoside ring (see Figure 1) or the CH_{3,4} site which binds the fucose to an apolar patch on the upper face of the ring. Both types of interaction have been identified in X-ray crystallographic investigations of complexes between proteins and methyl α -L-fucopyranoside (MeFuc).

structure	ΔE	$\Delta E + \text{ZPE}$	D_0
CH ₁ - π_a	0.00	0.00	-22.06
CH ₁ - π_b	2.70	1.49	-20.57
CH ₁ - π_c	2.96	1.73	-20.33
CH _{3,4} - π	3.96	2.41	-19.65
CH ₁ - π_d	3.07	3.62	-18.45
CH ₂ - π	5.42	4.63	-17.43
CH ₁ - π_e	7.66	5.98	-16.09
OH ₂ - π	9.22	7.61	-14.46

Table 1. Relative energies and binding energies in kJ mol⁻¹, of the eight lowest energy structures of MeFuc•Tol- d_8 calculated using the MO5-2X functional and a 6-31+G** basis set. The ‘ $\pi_a, b, \dots$ ’ notation identifies different orientations of the aromatic ring, which differ only by rotation in the aromatic plane.

Figure 2 compares the computed OH vibrational spectra of the uncomplexed carbohydrate (fig. 2a) with the experimental IRID OH spectrum of the complex, which populates two alternative

structures, or structural families, (fig. 2b) and the OH vibrational spectra associated with the lowest energy structures of MeFuc•Tol- d_8 , CH_1 - π (figs. 2c-e) and $CH_{3,4}$ - π (fig. 2f), calculated using the MO5-2X functional. They are all very similar although the CH - π interactions do lead to very minor changes in the spectral patterns. Apart perhaps, from the minimum energy structure, each one of them is in good correspondence with each of the experimental spectra (though this is *not* true for the OH spectra associated with any of the calculated higher energy structures). It is tempting to associate the two experimental spectra with CH_1 - π and $CH_{3,4}$ - π bonded structures.

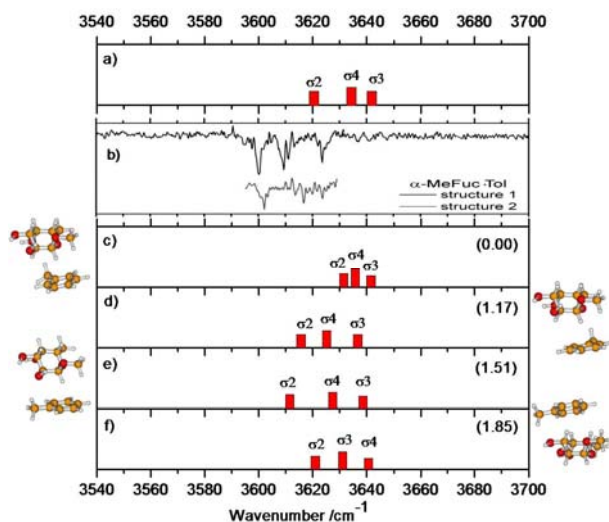


Figure 2. (a). Computed OH vibrational spectrum of methyl α -L-fucopyranoside; (b) IRID OH spectra (shown as 'dip' spectra) of the two populated conformers of MeFuc•Tol- d_8 and (c) – (f), the corresponding vibrational spectra and conformations of its lowest lying computed structures.

Similar comparisons conducted in the CH spectral region are presented in Figure 3, which presents the computed vibrational spectra of uncomplexed MeFuc and the four lowest energy MeFuc•Tol- d_8 structures, and the experimental IRID spectrum associated with the more strongly populated complex ('structure 1' in Fig. 2b). Disappointingly, all the spectra are qualitatively similar and this, when coupled with their complexity and the poor resolution and intensity of the experimental spectrum, prevents assignment of the complex to a particular structure or the vibrational assignment of its experimental spectrum. Comparisons between the computed spectra of each of the complex structures and the bare carbohydrate do however, reveal some interesting results. Each of the spectra shown in Figures 3c-3e is associated with a structure bound through a C_1 H- π interaction; when they are compared with the spectrum of bare MeFuc, 'blue shifts' can be identified in the respective C_1 -H bands ~ 35 cm^{-1} (c), ~ 64 cm^{-1} (d) and ~ 57 cm^{-1} (e) but none of the other bands is significantly displaced. Similarly, the C_3 -H and C_4 -H bands in spectrum (f), which is associated with a $C_{3,4}$ - π bound complex, are blue-shifted by ~ 30 cm^{-1} and ~ 66 cm^{-1} respectively, but not the other bands which are virtually unaffected. These large, selective displacements towards higher wavenumber are comparable in magnitude (and direction) with those calculated for similar CH - π bonded carbohydrate-toluene structures, using the dispersion corrected D-DFT method.^[3] Remarkably, blue shifts (about one third as large) are also predicted by calculations using the B3LYP functional although it is not capable of dealing with dispersive interactions.

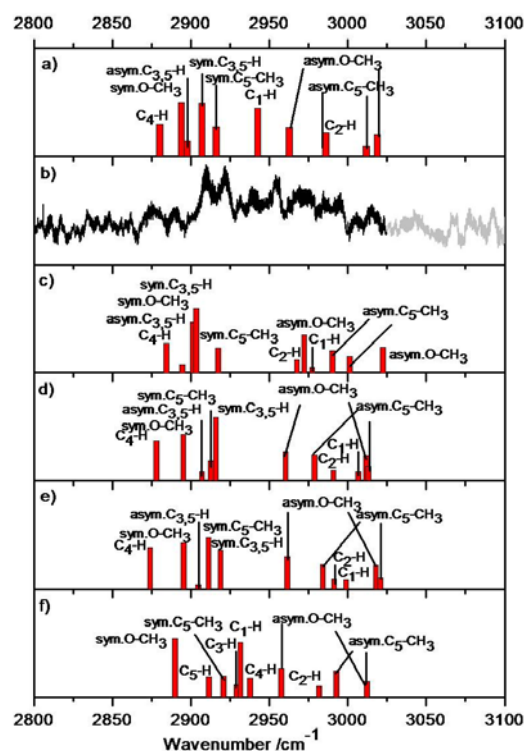


Figure 3. (a). Computed CH vibrational spectrum of methyl α -L-fucopyranoside; (b) IRID CH spectra of conformer 1 of MeFuc•Tol- d_8 ; (c) – (f) corresponding vibrational spectra of its lowest lying computed structures. (The weak features lying above ~ 3025 cm^{-1} are due to aromatic C-H bands associated with a small population of undeuterated toluene clusters).

Conclusions

The aromatic molecule is evidently bonded to the upper or the lower face of the pyranoside ring, through $CH_{3,4}$ - π (upper) or CH_1 - π (lower) interactions. The theoretical prediction of a strong blue shift in the CH bands associated with the CH - π interactions parallels^[3] the predictions made recently, on the basis of D-DFT calculations for other carbohydrate-toluene complexes,^[4] and encourages a further experimental pursuit based upon a simpler benchmark system, e.g. the xylose-toluene complex, methyl α/β -D-xylopyranoside-toluene- d_8 , which provides an analogue of glucose rather than galactose since all its OH groups adopt an equatorial orientation. If the methoxy group were fully deuterated, the number of CH bands would be almost halved to allow a considerable spectral simplification.

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Polymer coil-globule transition dynamics on the nanosecond to second time scale

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Introduction

Poly(N-isopropylacrylamide) (pNIPAm, Fig 1) undergoes a change in conformation from a hydrated random coil to a partially dehydrated globular structure when heated over the lower critical solution temperature (LCST). Aggregation of the globules due to hydrophobic interactions and intermolecular hydrogen bonds then leads to phase separation. Applications of this effect include thermally or pH switchable drug or DNA delivery and control of enzymatic activity.^[1]

This phase transition has been studied extensively; however, the *dynamics* of the transition has received only little attention so far. The temperature-jump (T-jump) induced coil-globule dynamics of cross-linked pNIPAm gel nanoparticles have been reported to occur on the 100 ns time scale,^[2] but dynamics of the single chain polymer phase transition are still in question.^[3]

We use laser-induced T-jumps and IR spectroscopy to follow the polymer collapse. We report evidence for fast local collapse of the polymer, but very slow phase separation in single chain pNIPAm samples. On the other hand, phase separation occurs on the 100 ms time scale at concentrations resulting in significant overlap of the polymers in the coil state.

Experimental

PNIPAm polymer ($M_n \sim 20,000$) was synthesized by standard methods, dissolved in D_2O and placed in a temperature-controlled IR cell with CaF_2 windows and 50 μm pathlength. The pNIPAm LCST was confirmed to be at 35°C by UV-vis spectroscopy. FTIR spectra were corrected for D_2O absorbance.

T-jumps were induced with visible laser light using co-dissolved heat transducers which absorb the laser energy and rapidly convert it to heat; here, either gold nanoparticles or the dye Basic Red were used. For measurements on the ns-time scale, a pulsed Nd:Yag laser (532 nm, 1 mJ) is used to heat the sample within 5 ns. The accessible time window in this case is limited to less than 1 ms due to cooling by thermal diffusion. For accessing longer time scales, an Ar-ion laser (1 W at 514.5 nm) was used. After a fast shutter turns on the laser beam within 1 ms, the irradiated volume warms up and reaches thermal quasi-equilibrium on the time scale of 10 ms, Fig. 3A, limited by the speed of thermal diffusion. In both cases, the IR absorbance of the polymer solution was followed using a tunable cw lead-salt diode IR laser and a fast MCT IR-detector (rise time 20 ns).

The size of the T-jump was determined from the absorbance change at 1584 cm^{-1} , which is due to the shift of the D_2O association band with temperature, whereas the polymer absorbance at this wavelength is small and shows no change with temperature (Fig. 1A); comparison with FTIR spectra of D_2O allows direct calibration of the temperature change.

Results – Temperature Dependent FTIR Spectra

FTIR spectra of the amide I' band, which is dominated by the amide-carbonyl stretch vibration, taken at incremental temperatures around the LCST, are shown in Fig. 1A. The position of this band responds strongly to changes of the polymer confor-

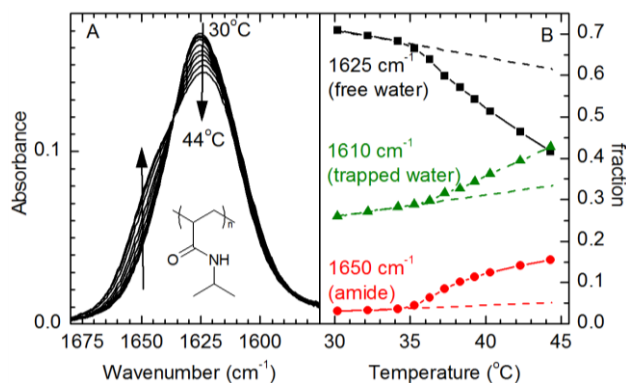


Fig 1. (A) FTIR spectra in the amide I' region of pNIPAm (10mg/ml in D_2O ; inset shows chemical structure, $n \sim 200$) at temperatures around the coil-globule transition (30–44°C), arrows indicate increasing temperature. (B) Fraction of the three component bands obtained from spectral deconvolution with pseudo-Voigt bands, see text for details.

mation. Below the LCST, pNIPAm exists as a hydrated coil, and the amide I' band is dominated by a band at 1625 cm^{-1} due to carbonyls hydrogen bonded (H-bonded) to water. Above the LCST, part of the amide I' band shifts to 1650 cm^{-1} due to intramolecular (amide-amide) H-bonds.^[4] Spectral deconvolution shows a significant contribution of a band at 1610 cm^{-1} , which can be ascribed to carbonyls bound to water molecules trapped in collapsed polymer sections. The fraction of carbonyls which are hydrated, form intramolecular H-bonds or are H-bonded to trapped water can be estimated from the band areas obtained from the fits, Fig. 1B. There is a small loss of freely hydrated carbonyls and increase of trapped water with increasing temperature even below the LCST, indicating the increased formation of locally collapsed polymer sections (see below). At the LCST, larger structural changes occur, including the formation of intermolecular H-bonds as the chains collapse and phase-separate.

Results - Nanosecond Dynamics

Fig. 2 shows time-dependent IR absorbance changes following a T-jump over the LCST. The size of the T-jump was determined from the signal at 1584 cm^{-1} , which is due to D_2O , as described above. The absorbance recovery at 1584 cm^{-1} on the 100 μs -time scale indicates cooling due to thermal diffusion. The absorbance at 1622 cm^{-1} , which has a contribution from the polymer, also decreases instantaneously within our time resolution and then remains constant in the window of ~ 1 ms accessible by our ns-setup, following the 1584 cm^{-1} absorbance changes exactly, Fig. 2. However, the absorbance change at 1622 cm^{-1} is larger than that at 1584 cm^{-1} , although D_2O has a smaller temperature-induced absorbance change at this wavelength; this shows that there is an instantaneous bleach of the polymer absorbance. The observed polymer absorbance change agrees well with that predicted from the temperature-dependent FTIR spectra by extrapolating the sub-LCST temperature dependence to temperatures above the LCST (dashed lines in Fig. 1B). As discussed in more detail below, this indicates that a T-jump-induced *local* collapse of PNIPAm occurs within a few ns.

On the other hand, the 1622 cm^{-1} absorbance change on the ns-time scale is smaller than the overall change predicted from the FTIR spectra. This shows that a second slower phase of relaxation, namely aggregation and phase separation, is too slow (slower than 1 ms) to be observable in these measurements.

Results – Millisecond Dynamics

Measurements on the time scale 10 ms to 5 s are shown in Fig. 3. At a concentration of 10 mg/ml, Fig. 3A, the results for 1584 cm^{-1} are identical to those obtained in the absence of polymer and thus provide a direct measure of the temperature after turning on cw irradiation. The LCST is reached after 10 ms; within 100 ms 90% of the final T-jump has been reached. At 1622 cm^{-1} , a larger absorbance decrease is found which is in full agreement with the results on the 10 ns-time scale (Fig. 2) when taking into account the slightly different experimental conditions. Again, the time dependence of the absorbance changes at the two wavelengths is identical, indicating that even in this longer time window no major structural changes occur after the initial local collapse. Additionally, we obtained the absorbance change at 1641 cm^{-1} , where the bleach of the D_2O absorbance is partly compensated by an absorbance increase of the polymer. These results are also fully compatible with local collapse only.

Completely different results are obtained at a concentration of 45 mg/ml, however (Fig. 3B). Here, the same instantaneous absorbance change due to local collapse is found (accounting for larger polymer absorbance at the higher concentration). After an induction period of 200 - 400 ms, however, significant changes of the absorbance at all wavelengths occur with a time constant of $\sim 500\text{ ms}$. The same results were obtained for concentrations in the range 30 - 100 mg/ml. Most interestingly, these absorbance changes cannot arise from structural changes of the polymer, since the polymer absorbance decreases both at 1622 cm^{-1} and 1641 cm^{-1} , whereas at 1584 cm^{-1} , where the polymer does not have any significant absorbance at all, it increases. These observations allow only one conclusion, namely that polymer is removed from the probed volume (decreasing amide I' absorbance) and is being replaced by water (increasing absorbance at 1584 cm^{-1}); the size of the observed effects is consistent with $\sim 15\%$ of the polymer being replaced by water.

Discussion

Cooperative hydration, which results in a “pearl-necklace” type chain conformation with locally collapsed polymer globules even below the LCST, has been suggested as the mechanism behind the pNIPAm coil-globule transition. Theoretical considerations predict an increase of the number and/or size of locally collapsed polymer sections with increasing temperature even below the LCST.^[5] This is fully confirmed by our FTIR spectra, which show a decreasing fraction of normally hydrated carbonyls and increasing number of carbonyls bound to trapped water with increasing temperature below the LCST (Fig. 1B).

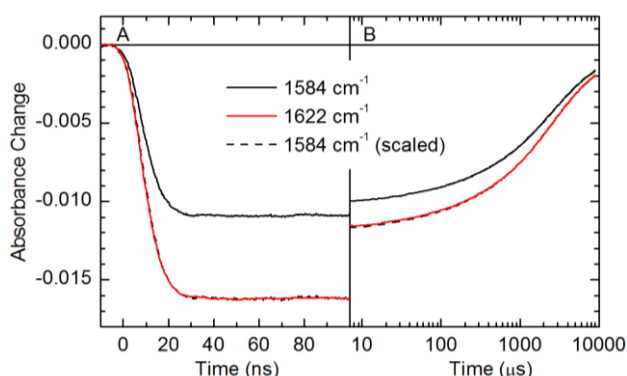


Fig 2. Absorbance changes of a pNIPAm sample in D_2O on the time scale 10 ns to 1 ms. (A) 20 mg/ml, T-jump from 32°C to 37.5°C ; (B) 7 mg/ml; T-jump from 32°C to 37°C .

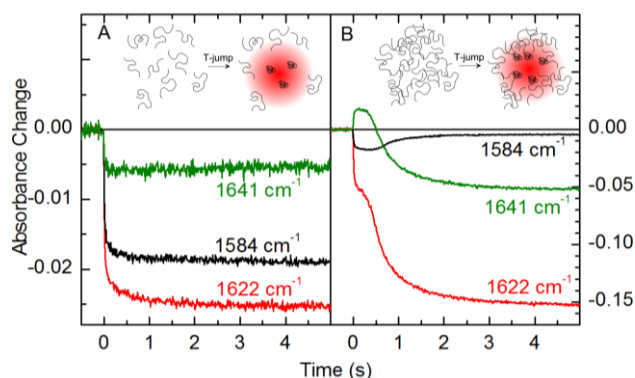


Fig 3. Absorbance changes on the time scale 10 ms to 5 s after a T-jump from 32°C to 40°C at different concentrations of pNIPAm: (A) 10 mg/ml. (B) 45 mg/ml.

The dynamics of the coil-globule transition of pNIPAm after a T-jump over the LCST is characterized by two phases. The first is very fast ($<10\text{ ns}$), and its spectral signature (not shown) corresponds to that expected from the extrapolation of the sub-LCST temperature dependence of the FTIR spectra to temperatures above the LCST. We conclude that even after a T-jump over the LCST, the polymer initially undergoes a similar local collapse as upon a temperature increase below the LCST, involving an increase of the number and/or size of collapsed polymer sections. Due to the local nature of this relaxation, it is not surprising that it proceeds on the nanosecond time scale.

At low concentrations, no further structural changes or phase separation occur up to at least 5 s. However, at higher concentration, phase separation leads to the removal of $\sim 15\%$ of the polymer chains from the heated volume on the 100 ms-time scale. The lowest concentration for this effect to occur is 30 mg/ml, corresponding to a mean polymer separation of $57\text{ \AA}$. This compares reasonably well with the radius of gyration of the coil polymer ($R_g \sim 15\text{ \AA}$), suggesting the onset of some overlap/entanglement between polymers. We propose that two conditions are required for polymer chains to undergo the phase separation on the 100 ms time scale observed here: (i) entangled polymer chains, so that the collapsing chains can provide a pulling force, and (ii) a temperature gradient to provide a directional preference for the pulling “force” of the collapsing polymers. This is schematically indicated in the insets to Fig. 3.

Conclusions

IR spectroscopy gives detailed insight into the mechanism and dynamics of the pNIPAm coil-globule transition. An increase of temperature results in fast local collapse of the polymer, followed by much slower aggregation and phase separation. Partial phase separation was observed on the 100 ms time scale, but only if the polymer chains are sufficiently entangled.

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Using tip enhanced femtosecond lasers to create graphite nanostructures on diamond

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Introduction

Graphene has become one of the most dynamic areas of condensed matter physics due to an unusual conflation of interesting fundamental physics and the very real possibility of important technological applications. Not only is it an excellent condensed-matter analogue for two-dimensional quantum electrodynamics[1, 2], but it is also a zero band gap semiconductor with room temperature ballistic conduction and electron mobilities at least two orders of magnitude greater than in silicon.

Theoretical predictions of the transport properties of nanoscale graphene and graphite are particularly intriguing. Thin ribbons with largely zig-zag edges are predicted to be metallic and order magnetically, they are reported to be magnetic-resistive and even half metallic[3, 4] and by mixing in edge-states it is possible to open up the band-gap necessary to create electronic devices. To do this it is necessary to be able to control orientation of the ribbon with respect to the crystal structure.

Although much of the current interest is focused on graphene it now appears that some of its properties, such as presence of Dirac Fermions[5, 6] and ballistic conduction[7], are also present in graphite [8]. But research and technological application is hampered by a lack of sample. Although it is possible to produce high quality crystals of graphene of up to 100 μm through mechanical cleavage these methods are difficult to scale up and many groups are looking for alternatives. Our aim was to see if it was possible to grow thin layers of graphite through femtosecond laser irradiation of an atomically flat diamond (111) surface. Although we have demonstrated this and made progress in understanding the mechanisms of the diamond – graphite transformation the control of the thickness and lateral dimension has proved difficult. The thickness is controlled by multi-photon absorption and has a non-linear dependency with both wavelength and laser power and the lateral dimension is limited by the optical diffraction limit.

The aim of this research was to use tip enhancement to focus and amplify light intensity under the tip of a conducting atomic force microscope so that we can create structures on the surface of diamond smaller than the wavelength of laser.

To do this we made an application to use the femtosecond laser (UFL2) from the loan pool to build a prototype of a tip enhanced femtosecond laser and use it to create graphite nanostructures for transport measurements.

Femtosecond irradiation of diamond

Heating the diamond (111) surface to temperatures of 2000K will transform it to graphite. This can be done in a furnace or with a laser with a pulse width of a nanosecond or more. However when the pulse length is reduced to the order of a hundred femtoseconds then there is no possibility for the electron-hole plasma reaching equilibrium with the lattice on the timescale of the laser pulse and recent theory and experiment indicate that the transformation is athermal and driven by the plasma itself[9, 10]. Our initial non-tip enhanced experiments appear to confirm this; showing the formation of small blisters of graphite which, are crystallographically aligned with the underlying diamond lattice. This means it should be possible to create nanostructures with controlled amounts of zig-zag and arm-chair edge-states and hence control the band-gap of a nano-graphitic ribbon.

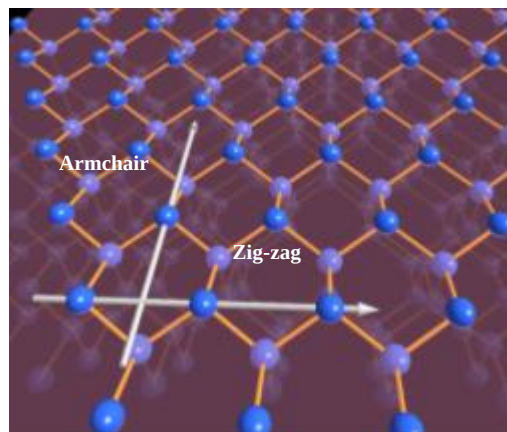


Figure 1: The (111) surface of diamond showing the puckered graphite like arrangement of the atoms. The two arrows show the armchair edge direction $\langle 1\bar{1}1 \rangle$ or the zig-zag edge $\langle 011 \rangle$.

In our previous experiments using the CLF lasers we were able to irradiate an atomically flat diamond (111) surface with a 120fs laser pulse with wavelengths between 200 and 800nm. Diamond has an indirect band-gap of 5.4eV, but fitting of the transmission as a function of power shows that it is the direct band gap of 7.3eV that controls the production of electron-hole pairs[11]. As a consequence at all the wavelengths we are using the absorption processes are multi-photon and have a non-linear dependency with laser power. At 200 and 266nm we are firmly in the two-photon absorption region and we find that at above the graphitization threshold of 0.1 - 0.6 J/cm² we get a layer of

graphite with a thickness that scales with the absorption depth but with a relatively small crystallite size. But at 800nm we get well defined graphitic triangular crystals that are aligned with respect to the underlying diamond lattice and have a crystallite size in excess of 80nm (Figure 2). These grow from relatively sparse nucleation sites, implying that there are defects in the diamond that have levels in the band gap to act as generators of electron-hole pairs.

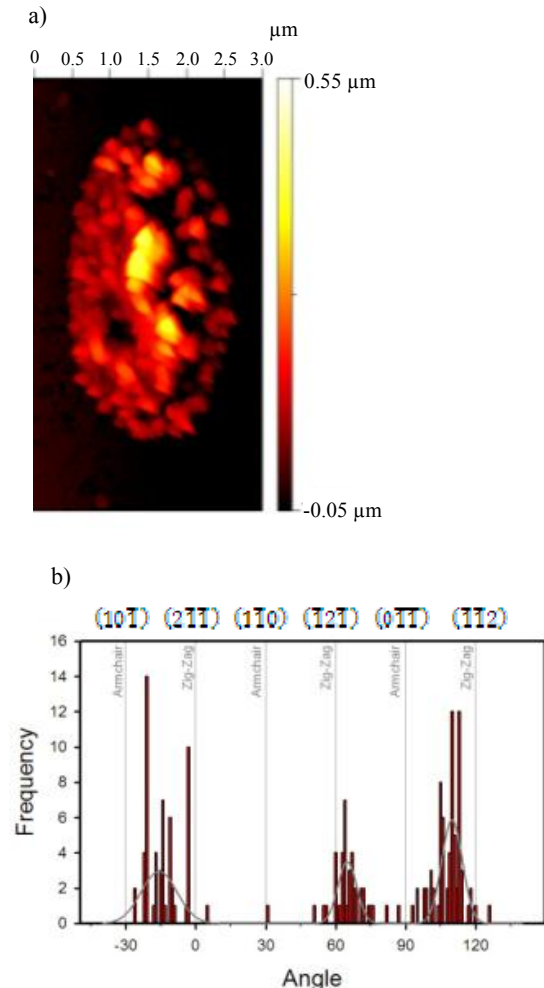


Figure 2: a) An Atomic Force Microscope height image showing triangular graphite crystals ~200nm across on the (111) surface of diamond. The spot was created with an 800nm 120fs laser pulse with an integrated energy of 3μJ. b) A plot of the distribution of surfaces in the edges of the triangles. The centres of the distributions are at -15 ± 2 , 65 ± 1 and 119.8 ± 0.7 degrees and so the triangles are not equilateral but are clearly closer to zig-zag than armchair. The orientation of the triangles is consistent across the whole spot and between spots implying that they are related to the underlying crystal.

Tip Enhancement

A metal tip brought close to a surface can create several orders of magnitude enhancement of the electric field through a combination of electrostatics and surface plasmon excitations that resonate with laser frequency. There have been rapid developments in techniques that use this tip enhancement for spectroscopy (Tip Enhanced Raman Spectroscopy, TERS) and optical microscopy (Scanning near-field Optical Microscopy, SNOM or aperture less SNOM, ASNOM)[12]. And there have been a few attempts to use it to modify surfaces by ablating metals or glass to create pits and lines and there have been measurements to modify the conductivity of hydrogen passivated silicon surfaces[13-18]. Although this idea has been in the literature for around 10 years it is only in the last 2 years that the first attempts have been made with femtosecond lasers.

This is clearly important for materials where the transformation processes is driven by high order multi-photon processes.

So far no-one has reported a method to transform a material from an insulator into a conductor or semiconductor. If we are able to do this on diamond or silicon carbide we would be able to write graphitic nano-electronic devices directly onto a surface with dimensions equal to those possible using state-of-the-art lithographic techniques in the electronics industry

The experimental set-up

We focused the light from the UFL2 laser onto test surface and the tip of silicon tip the AFM. By using a highly attenuated beam it was possible to view the laser spot using the camera and optics of the AFM.

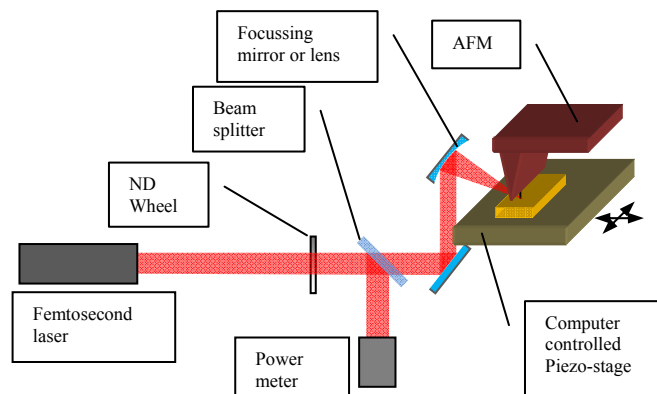


Figure 3: A schematic of the laser set-up that we envisage for the prototype.

All the initial set-up was done using a silicon wafer as the substrate. When the tip was advanced to the surface of the silicon the enhancement of the laser was immediately obvious as the tip glows brightly. We then set AFM to track a given path at a given speed along the surface in contact mode, whilst the laser was pulsed at up to 1000 pulses using 800nm wavelength and pulse width of 120fs.

Results and discussion

During this set-up phase we found it possible to make sub-diffraction limit structures on the silicon with relative ease (figure 4). Switching to gold coated tips created an enhancement of between $\times 10 - \times 100$ tip enhancement because of the much more favorable Plasmon frequencies.

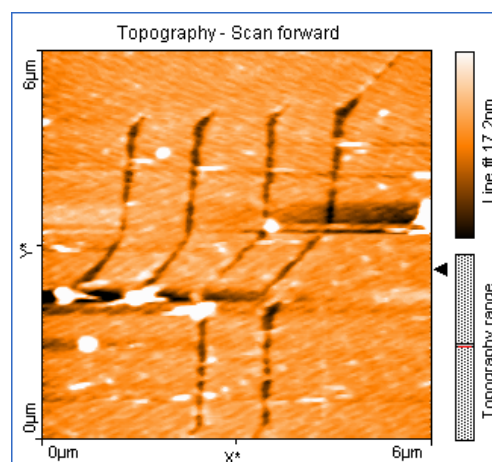


Figure 4: An AFM scan of structures written onto a silicon surface, using 800nm laser with a pulse width of 120fs running at 1000Hz. This is using a gold tipped AFM moving at speeds ranging 4 μm/s to 256 μm/s. The structures are trenches approximately 100nm across.

However, we were not able to replicate this work on the diamond. This is probably partly due to the fact that the enhancement that you can achieve on a non-conducting surface is considerable smaller than for a conducting surface. We were hoping to be able to excite electrons over the band gap to promote conductivity, but at 800nm we are relying on very high order multi-phonon processes. We were hoping to be able to use shorter wavelength laser light, but this proved impossible in the timescales available.

Conclusions

Although we were able to achieve tip enhanced structural changes on a conducting surface we were not able to do this on the non-conducting diamond surface. It may still be possible to do this at shorter wavelengths.

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Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing

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Introduction

Fluorescence based sensing uses media doped with rare earth or transition metal ions in which the spectrum of the emitted fluorescence or the excited state decay time (τ) depends on the external environment^[1,2]. The strong temperature dependence of τ has enabled laboratory resolutions of $<1^\circ\text{C}$ to be demonstrated^[3]. Its weak strain dependence can also be exploited in conjunction with the strain independence of the intensity ratio of thermally linked spectral lines to yield strain sensitivities of ~ 250 microstrain ($\mu\epsilon$)^[4]. In practice though, for the purposes of temperature sensing, the temperature to strain cross-sensitivity for most dopants is sufficiently small that even the application of a breaking strain only produces a temperature uncertainty of a few degrees Celsius. Existing fluorescence based sensors employ single-photon excitation and are thus point sensors in which a small sample of active material is attached to the end of a conventional optical fibre. The optical fibre plays no role other than to transmit the excitation light to the sample and to return the resulting fluorescence to a remote detector. Quasi-distributed sensing of discrete points using single-photon excited fluorescence (SPF) is only achievable by a network of such sensors. Currently, continuously distributed fibre sensing is achievable by optical time domain reflectometry of spontaneous and stimulated scattering, but these sensors frequently have limited temporal and spatial resolutions.

Distributed optical fibre sensing based on two-photon excited fluorescence

The authors have previously shown^[5-7] that two-photon excited fluorescence (TPF) may be exploited to achieve true distributed sensing. TPF is generated in an ionic medium by the simultaneous absorption of pairs of photons without the involvement of a real intermediate energy level. The sum of the photon energies in each pair matches the energy gap from the ground state to the higher energy level from which the ions relax with the eventual emission of fluorescence photons. With the TPF intensity being effectively proportional to the square of the total local photon flux, an enhanced fluorescence signal is produced where two laser pulses overlap. If the two pulses are counter-propagating in a doped optical fibre, their meeting point can be scanned along the length of the fibre by introducing a variable time delay between them. The resulting fluorescence flash from the overlap, usually in the visible or near infrared, is then transmitted along the fibre and detected and analysed at one end, Figure 1. With the length of the sensed position depending only on the pulse duration, there is the potential for high spatial resolution, while the total length of a fibre designed for two-photon absorption is intentionally transparent at the single-photon wavelength and is not normally limited by attenuation. Distributed TPF based sensing is equally applicable to mono- and multi-mode doped fibres allowing the technique to be extended to single crystal fibres such as sapphire with core diameters of 100s μm . Although modal dispersion of the excitation pulses will now reduce the spatial resolution, this will be more than compensated for in specific applications by their tolerance to much higher temperatures ($\sim 1500^\circ\text{C}$) than glass fibres. Thus a TPF based sensor is expected to simultaneously

possess high temporal (seconds), spatial (down to mm) and temperature ($\sim 1^\circ\text{C}$) resolutions.

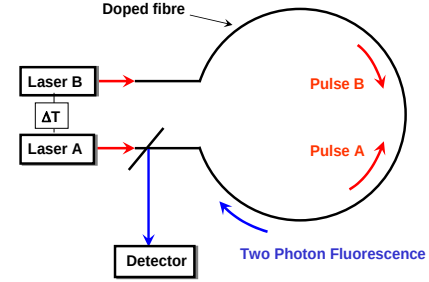


Figure 1. Schematic diagram of a distributed sensor based on time-correlated TPF in a doped optical fibre. Counter-propagating pulses (A) and (B) meet at a location depending on their relative time delay, ΔT . The TPF generated at their overlap is guided along the fibre and detected.

Nonlinear spectroscopy

The aim of this work was to obtain the two-photon absorption spectrum for glass and crystal doped with various lanthanide and transition metal ions. Without such information, the feasibility of a distributed fibre sensor based on TPF cannot be assessed. The existing spectroscopic literature for these materials is mainly concerned with single-photon excitation linked to applications as laser media or fluorescence point sensors. Even for the latter, the published temperature and strain characteristics of the fluorescence lifetimes and spectra are generally not relevant to the approach described here as they have only been studied for the near-IR emission lines that arise from single-photon excitation and not visible TPF.

Measurement of the two-photon absorption cross-section

Direct measurement of the two-photon absorption cross-section (δ) is challenging because of the small absorption which has to be detected as well as knowing the ion concentration (N_i). To circumvent these difficulties, the TPF yield can be compared, under identical conditions, with that for SPF allowing δ to be deduced in terms of σ , the single-photon absorption cross-section. Although the unknown N_i can now be eliminated, measurement of the fluorescence signals is non-trivial due to their vastly different magnitudes.

It is relatively straightforward to show for sample thicknesses ≤ 0.1 of an absorption length ($1/\sigma N_i$), that under excitation by nanosecond Gaussian pulses from a Q-switched laser,

$$\delta = \frac{1}{2} \sqrt{\frac{\pi}{2 \ln 2}} \frac{A'' t_p'' hc}{\lambda''} \left(\frac{E'}{E''^2} \right) \left(\frac{P''(t)}{P'(t)} \right) \sigma \quad (1)$$

where $P'(t)$ and $P''(t)$ are the SPF and TPF powers respectively, E' and E'' are the energies of the SPF and TPF excitation pulses respectively, λ'' and t_p'' are the wavelength and FWHM duration of the TPF excitation pulses, and A'' is the area of the TPF excitation pulses in the sample. The quantum efficiencies for the two excitation schemes cancel when the same transition is involved in both cases, but accurate knowledge of E' and E'' , t_p'

and A'' at λ'' is still required to evaluate (1).

The two-photon absorption spectrum of ruby

The two-photon absorption spectrum of ruby (Cr^{3+} :sapphire) is described here as an example of the type of measurement made. It helps complete the quantitative investigation of this important crystal which has been at the forefront of optoelectronics for over half a century as well as being the first practical laser medium. Related experiments on doped glass are outlined in the accompanying report – “Distributed sensing by time-correlated two-photon excited fluorescence in rare earth doped optical fibres” by I S Ruddock et al.

Transition metal ions offer broad absorption bands in the visible region. Cr is the most studied and the ‘R-line’ emission of ruby has been used in SPF based point sensors of pressure, strain and temperature by detecting the spectral shift of the fluorescent R-lines [1] and the change in the fluorescence decay time [2]. Sapphire fibres can be produced by laser heated miniature pedestal growth [8] and by extrusion [9].

The strong R-line fluorescence (R_1 , 694.3 nm and R_2 , 692.9 nm) is due to decay from the metastable 2E state (the lowest lying component in the free ion’s 2G first excited state) back to the 4A_2 ground state (the lowest lying component in the free ion’s 4F ground state), Figure 2. The metastable 2E state is split into $\bar{E}$ and $2\bar{A}$ states with an energy gap of $\sim 29\text{ cm}^{-1}$ by the nearly octahedral symmetry of the sapphire (Al_2O_3) host. The fluorescence decay time (τ) decreases from $\sim 3.5\text{ ms}$ at room temperature to $\sim 45\text{ }\mu\text{s}$ at $650\text{ }^\circ\text{C}$ and the R-lines shift slightly to longer wavelengths with increasing splitting, thus providing parameters for sensing purposes [10].

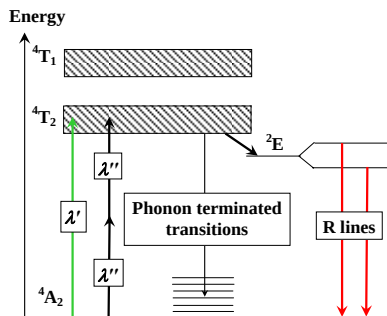


Figure 2. Simplified energy level diagram for ruby showing single-photon (λ') and two-photon (λ'') excitation.

The measurements reported here were performed on a sample cut from a standard a-axis ruby rod. Excitation was provided by two Continuum pulsed systems: a Q-switched and frequency doubled Nd:YAG laser and the Laser Loan Pool’s NSL3 optical parametric oscillator (OPO), both producing pulses with energies of 10s mJ and durations of ns. For light polarised along the ordinary and extraordinary axes of the crystal, the TPF and SPF signals were measured at $1.064\text{ }\mu\text{m}$ and 532 nm to determine discrete values of δ at $1.064\text{ }\mu\text{m}$ using equation (1) along with the appropriate values of σ at 532 nm [11].

The wavelength dependence of δ over the 800–1200 nm range, i.e. the two-photon absorption spectrum, was obtained for the first time by recording the two-photon excitation spectra for the R-lines using the OPO’s idler output and then converting them to absorption by normalising against (i) the square of the wavelength dependence of the OPO’s photon flux, and (ii) the wavelength dependence of the fluorescence quantum efficiency [12], and finally scaling with the two values of δ already calculated for $1.064\text{ }\mu\text{m}$, Figure 3.

The maximum values of δ observed were $5.9 \times 10^{-3}\text{ GM}$ ($1\text{ GM} = 1 \times 10^{-50}\text{ cm}^4\text{ s ion}^{-1}\text{ photon}^{-1}$) for e-polarisation and $4.6 \times 10^{-3}\text{ GM}$ for o-polarisation at 840 nm . The overall uncertainty is

$\pm 40\%$, due primarily to the precision of the absolute quantities in equation (1). The random uncertainty in the underlying two-photon absorption spectrum is $\pm 20\%$. In general, the two-photon and single-photon absorption spectra (inset in Figure 3) are similar. There are two broad bands of unequal strength corresponding to absorption from the ground state into 4T_1 and 4T_2 showing the same polarisation dependence. The longer wavelength peaks in λ'' at $\sim 1100\text{ nm}$ agree well with those at $\sim 2 \times 550\text{ nm}$ in λ' . The shorter wavelength peaks at $\sim 840\text{ nm}$ are offset by 30 nm from $\sim 2 \times 405\text{ nm}$.

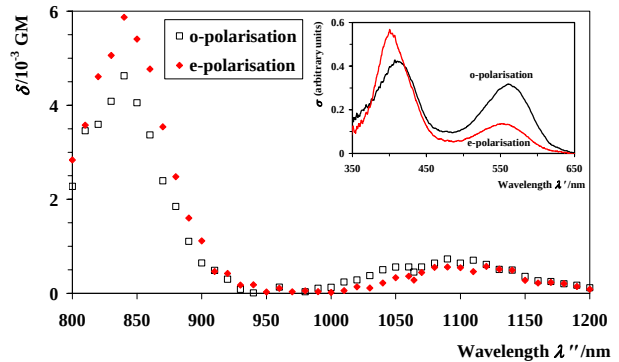


Figure 3. The two-photon absorption spectrum of ruby for ordinary and extraordinary polarised light. The inset shows the normalised single-photon absorption spectrum.

Conclusions

Knowledge of δ for any fluorescent crystal or glass enables their use in a TPF based distributed optical fibre sensor to be assessed in the light of available lasers and other measurable and predictable parameters. Using the values reported here and the theory in [7], it can be shown that $>10^4$ fluorescence photons are detectable at one end of a ruby fibre of Cr density $\sim 10^{17}\text{ cm}^{-3}$ (corresponding to a fluorescence self-absorption length of 10 m), diameter $100\text{ }\mu\text{m}$, quantum efficiency ~ 0.9 and internal numerical aperture 0.38 (unclad sapphire) by pairs of counter-propagating 850 nm pulses of energy $1\text{ }\mu\text{J}$ and duration 1 ns . In four-level media unlike ruby, self-absorption does not restrict the dopant density and the fibre is transparent for the fluorescence as well as the excitation pulses. Even with a decrease in the quantum efficiency at elevated temperatures, there is a sufficiently large fluorescence flux to enable τ to be measured rapidly and with precision.

Acknowledgements

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Distributed sensing by time-correlated two-photon excited fluorescence in rare earth doped optical fibres

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Introduction

Continuously distributed fibre sensing is normally achieved by optical time domain reflectometry of spontaneous and stimulated scattering, but these sensors frequently have limited temporal and spatial resolutions. Alternatively, the environmental dependence of the fluorescence spectrum or the excited state decay time (τ) for glass or crystal media doped with rare earth or transition metal ions can be exploited^[1,2]. Additional detail on distributed sensing based on two-photon excited fluorescence (TPF) is contained in the accompanying report – “Nonlinear spectroscopy of doped glass and crystal for applications in distributed fibre sensing” by I. S. Ruddock et al., along with related experiments on the measurement of the two-photon absorption cross-section.

The basic approach of distributed sensing by fluorescence is that counter-propagating laser pulses in a doped fibre generate enhanced TPF at their overlap due to the absorption at this point being proportional to the square of the total local photon flux. The TPF is guided by the fibre to a detector at one end, while the overlap position can scanned along the fibre by means of a variable delay between the excitation pulses. Experiments and simulations show that a TPF based sensor is expected to simultaneously possess high temporal (seconds), spatial (down to mm) and temperature ($\sim 1^\circ\text{C}$) resolutions.

Despite the two-photon absorption cross-section (δ) being of the order of 1 GM ($= 1 \times 10^{-50}\text{ cm}^4\cdot\text{s}\cdot\text{ion}^{-1}\cdot\text{photon}^{-1}$), sufficiently large TPF powers can be generated with relatively modest diode laser sources. With the spatial resolution depending only on the extent of the pulses' overlap, i.e. their second-order cross-correlation, even 'long' ns pulses still correspond to spatial resolutions of 10s cm. Surprisingly however, the use of such pulses does not affect the amount of TPF generated - the TPF produced at the overlap depends only on the energies of the excitation pulses and not their intensities or temporal profiles^[3]. This is effectively due to a trade-off between the intensity of the stationary overlap region and the number of ions illuminated, in contrast to what would happen if TPF is generated by transient pulses away from the overlap. As TPF based sensing is equally applicable to mono- and multi-mode doped fibres, the technique can be extended to single crystal fibres with core diameters of 100s μm . Although modal dispersion of the excitation pulses will now reduce the spatial resolution, this will be more than compensated for in specific applications by their tolerance to much higher temperatures ($\sim 1500^\circ\text{C}$) than glass fibres.

Two-photon excitation spectroscopy

The identification of potential TPF based sensing media was performed by exciting bulk samples of rare earth doped fluoride and silicate glass with the tuneable near-infrared idler and the visible signal outputs from the Laser Loan Pool's NSL3 optical parametric oscillator (OPO). Single- and two-photon excitation

spectra were recorded by monitoring the fluorescence from various emission lines in each sample. Two-photon excitation spectra show the optimum wavelengths for excitation, but must be used in conjunction with appropriate single-photon excitation spectra to ensure that true TPF is being observed and not two-step fluorescence; in two-step fluorescence, a real intermediate energy level is excited by the first photon from which the second photon promotes the ion to the upper energy level where it relaxes with the eventual emission of a fluorescence photon. In the most general model of the sensor, the counter-propagating pulses can separately produce background TPF of the same wavelength as the TPF flash from their overlap. Detailed spectral information is necessary for choosing non-degenerate excitation wavelengths that only produce TPF at the overlap of the pulses and not individually. A fuller discussion of the choice of pulse wavelengths, energies and durations, and the impact of fibre length is given in reference [3]. Also, by comparing the fluorescence yield for single-photon excited fluorescence with that for TPF, δ can be determined^[4].

Two-photon and two-step excited fluorescence was observed in praseodymium, samarium, europium, gadolinium, terbium, dysprosium, holmium and thulium^[5], Figure 1. From this list, praseodymium was selected for detailed investigation in single-mode fibre geometry on account of its convenient excitation and TPF wavelengths and its potentially negligible fluorescence self-absorption due to the transitions of interest near 600 nm terminating above the ground state.

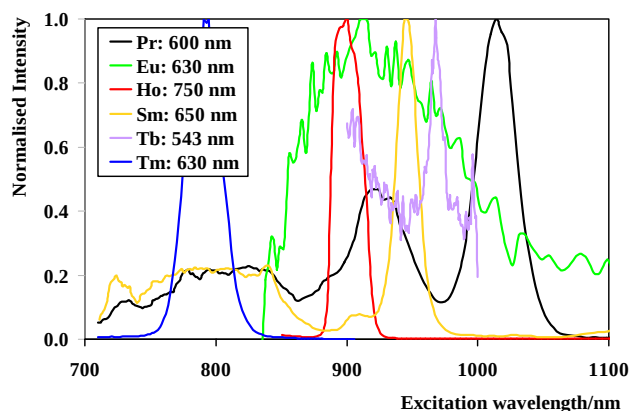


Figure 1. Two-photon and two-step excited excitation spectra for a range of rare earth doped glasses of different composition. The corresponding emission wavelengths are indicated in the legend. For praseodymium (black line), note the TPF excitation peak at $\sim 930\text{ nm}$ and the adjacent two-step excited fluorescence peak at $\sim 1014\text{ nm}$.

TPF in praseodymium doped fibre

Figure 2 shows the TPF spectrum at ~ 600 nm from 10 m of praseodymium doped single-mode fibre. The OPO's idler at 930 nm excited the ions in the vicinity of the 3P manifold centred on $\sim 2100\text{ cm}^{-1}$ above the 3H_4 ground state. The principal emission contains lines at ~ 595 nm, ~ 620 nm and ~ 635 nm. These are a superposition of transitions from 3P_0 and the lower lying 1D_2 , which is populated radiationlessly from the former.

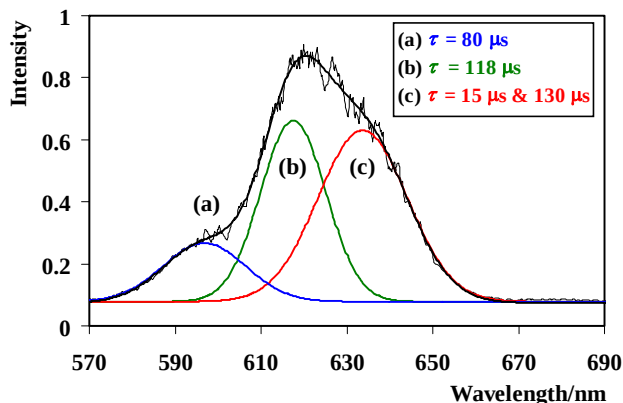


Figure 2. The TPF spectrum for praseodymium doped silica fibre showing the composite emission line at ~ 600 nm, the de-convolved components and their respective room temperature decay times, τ .

By comparing the effects of self-absorption in short and long lengths of fibre, the tentative identification of the components is that the 595 nm line corresponds to $^1D_2 \rightarrow ^3H_4$ but those at 620 nm and 635 nm are $^3P_0 \rightarrow ^3H_6$ and $^3P_0 \rightarrow ^3F_2$ respectively, Figure 3. As neither of these two longer wavelength TPF transitions terminate on the ion's 3H_4 ground state, they are suitable for distributed sensing over long paths.

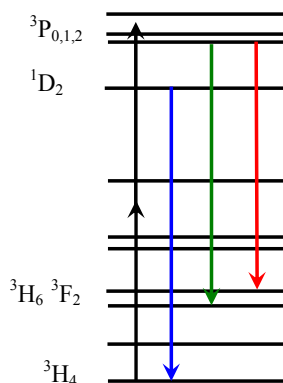


Figure 3. The energy level structure of praseodymium showing the fluorescence transitions following two-photon excitation at 930 nm.

The room temperature values of the fluorescence decay times for the three transitions are listed in the inset in Figure 2. The room temperature decay curve for the 620 nm emission is shown in Figure 4 along with its temperature dependence in the inset. With only 5000 data points, a 5% precision in the fitted lifetime is obtained even under two-photon excitation. This indicates that good temperature and temporal resolutions and short acquisition times will be achievable when a high repetition rate source (~ 1000 Hz) is employed in a prototype sensor instead of the 10 Hz OPO used here. Preliminary measurements of the time development of the TPF produced by degenerate counter-propagating pulses at 930 nm suggest that enhanced fluorescence (the TPF flash) is indeed being

generated at their overlap as predicted by the authors' theoretical model [6].

Conclusions

The use of the Laser Loan Pool's NSL3 OPO for investigating the two-photon excited fluorescence spectroscopy of rare earth doped glass and optical fibre has been summarised. New transitions in the visible suitable for distributed optical fibre sensing have been identified. Matched single-photon and two-photon excitation spectra for the same transitions have been recorded for certain dopants and their forthcoming analysis will enable the two-photon absorption cross-sections to be determined for these specific ion-glass combinations for the first time.

Acknowledgements

The loan of the NSL3 OPO is gratefully acknowledged by the authors. Craig Dalzell acknowledges support by an Engineering and Physical Science Research Council postgraduate studentship. The authors also thank OFS, Denmark for the supply of R31501-Pr fibre.

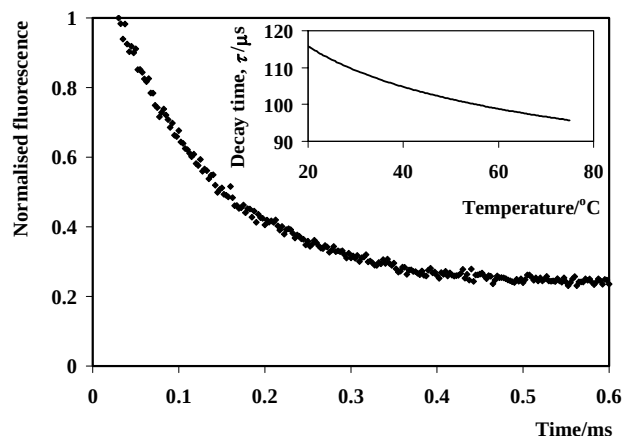


Figure 4. The room temperature decay of the 620 nm fluorescence from praseodymium doped fibre; $\tau = 118\text{ }\mu\text{s}$. The inset shows the temperature dependence of τ .

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Monochromatised XUV Beamline for ultrafast Time-Resolved ARPES

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Introduction

The short pulses of coherent XUV radiation produced through high harmonic generation have enabled the study of ultrafast electron dynamics in atomic systems and simple molecules. The Artemis facility now aims to exploit these XUV pulses to investigate ultrafast dynamics in experiments spanning gas-phase chemistry of polyatomic molecules and condensed-matter physics of complex materials

We have built an XUV beamline that delivers short pulses of monochromatised XUV, produced through high harmonic generation, to an end-station optimized for photo-emission experiments on condensed matter. We have shown that the beamline can achieve 250 meV energy resolution and 30 fs temporal resolution, enabling us to carry out first measurements of time-resolved photoemission with XUV pulses.

The core source of the Artemis facility is a Ti:Sa CPA laser system with two amplifier stages delivering 14 mJ/pulse, 30 fs, 800 nm pulses at 1 kHz repetition rate. The output beam is divided in three branches: 10 mJ allocated to a high power OPA (1 mJ @ 1.3 μm output) to cover the wavelength region 200 nm – 15 μm , 1 mJ for short pulse (7 fs) by hollow fiber compression and the rest for other pump applications. All the beams are Carrier Envelope Phase stabilized and intrinsically synchronized. Any of the beamlines can be used to generate XUV pulses through high harmonic generation, or used as synchronized pump and probe pulses.

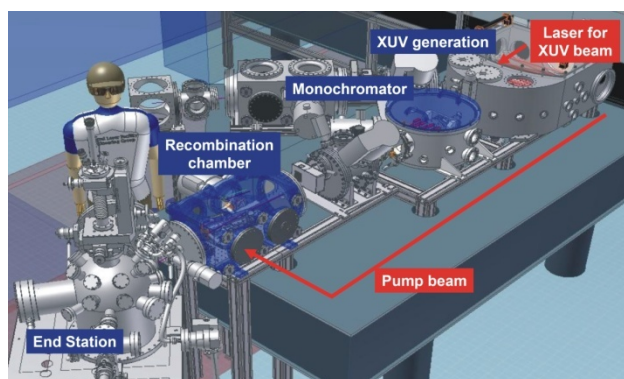


Figure 1: Layout of the XUV beamline including the Harmonic generation chamber, the monochromator chamber to select the photon energy, the recombination chamber for pump-probe experiment and the experimental chamber for time resolved ARPES.

The principal elements of the XUV beamline are the HHG chamber to generate short wavelength pulses, the monochromator to select the photon energy, the recombination chamber to align the pump/probe beams and the End-Station to perform the photoemission experiment (see Figure 1). All the optical elements are mounted on the optical bunch and the

vacuum chambers are isolated to minimize the mechanical vibrations.

High Harmonic Generation

High Harmonic Generation is a well established technique to produce short wavelength (12 nm, 100 eV) photons using high peak power tabletop laser sources. In a vacuum chamber, a lens focuses the drive laser beam onto a pulsed (1 kHz) gas jet synchronized with the laser system. After the interaction region the harmonics propagate collinearly to the incident beam with a smaller divergence angle (Figure 2a). During the laser-atom interaction, the Coulomb potential of the atom is deformed by the laser field allowing the tunneling of an outer electron in the vacuum. When the laser field changes polarity the electron is reaccelerated towards the ion. Then the electron recombines with the ion emitting an XUV photon (Figure 2b). For symmetry reasons only odd harmonics are generated.

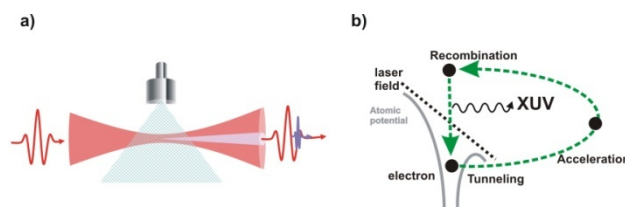


Figure 2: a) Sketch of the harmonic generation process. A pulsed laser with high peak power is focused on a gas jet. The harmonics propagate collinearly to the incident beam with a smaller divergence. b) Three steps model of the High Harmonic Generation: atomic ionization, electron acceleration and electron recombination generating the XUV photon.

The harmonic generation efficiency depends on the phase matching conditions at the interaction region and it is controlled by the drive beam diameter, the focal lens and the pulse energy, as well as gas pressure, length of interaction region and the position of the laser focus relative to the gas-jet. On a daily basis our conditions are 8 mm, 50 cm, 800 microJ respectively and we use 2 bar of Ar in a ~5mm long interaction region. The highest photon energy reachable depends on the gas used (Ar, Ne) and the wavelength (800 nm – 1.3 μm) of the drive beam.

Monochromator

The monochromator was developed in collaboration with LUXOR laboratory [1]. It was designed to select one single harmonic from the spectrum and preserve the harmonic pulse duration (~10fs). A first toroidal mirror collimates the incident beam and deflects it onto a plane diffraction grating. The diffracted light is refocused by a second toroidal mirror on to the exit slit (Figure 3a). The grating is used in conical illumination to limit the time spread of the pulses. Four gratings are mounted on a translation stage to cover the energy range 20-

100eV with resolving powers corresponding to either 10 fs or 50 fs pulse duration.

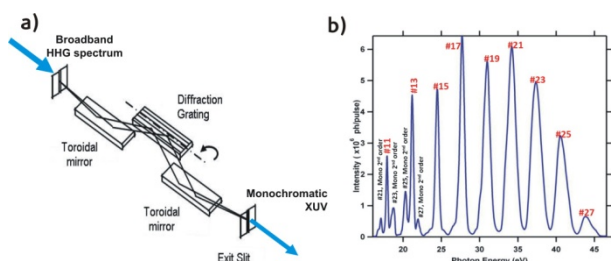


Figure 3: a) Layout of the monochromator including two toroidal mirrors to collimate the XUV on the grating and to refocus the beam on the exit slit after diffraction. b) Example of Ar harmonic yield measured after the exit slit.

The transmitted XUV intensity is measured by inserting a calibrated channeltron in the beam after the exit slit. In Figure 3 we present an example of harmonic yield obtained by rotating the grating and converting the rotation angle into photon energy.

Recombination chamber

After the monochromator exit slit the XUV beam diverges and is reimaged on the sample with a toroidal mirror in the recombination chamber. The gold coated toroidal mirror operates in 2f geometry at grazing incidence (4°). The 5 axis motorized mirror mount allows fine control of the spot size and shape on the sample. The pump beam (200 nm - 15 μ m) travels on the side of the beamline and is focused on the sample with a lens mounted on the optical table. In the recombination chamber the XUV beam goes through a holey mirror and the pump beam is reflected at 90° by this mirror to operate in collinear mode. A mirror mounted on a linear drive deflects temporally both beams to find the spatial and temporal overlap at the focal point (in air). The pressure gradient between the recombination chamber and the UHV chamber is maintained by a 2 mm diameter pin hole mounted on an XY translation.

Experimental Chamber

The experimental end-station is a fully equipped UHV chamber with a mu-metal shielding. The sample is mounted on a 5 axis manipulator with He cooling down to 14K. A load-lock allows fast transfer of the sample into the main chamber. After in-situ cleaving the surface quality is controlled with a LEED analyzer. The photoemission is performed with a hemispherical analyzer (PHOIBOS 100) where both angle and energy are resolved simultaneously. The analyzer resolution is better than 25 meV measured on Ag Fermi edge at low temperature. Depending on the electron optics mode, electrons are collected with high angular resolution ($\pm 0.1^\circ$) or over a large angular dispersion ($\pm 13^\circ$). A He lamp is available for off line sample characterization.

Energy and Time Resolution

In order to characterize the beamline overall energy resolution we present in Figure 4a the TaS₂ Fermi edge measured at room temperature using the monochromatized 17th harmonic. The red curve is a fitting of the data with a Fermi-Dirac function convolved to a 250 meV width Gaussian function. The XUV beam was monochromatized with the highest resolution grating ($E/\Delta E = 100$) and the exit slit set at 100 μ m. The photoemission energy resolution achieved (250 meV) is limited by the monochromator resolving power, which is designed to preserve the pulse duration of the ultrafast harmonics (~ 10 fs).

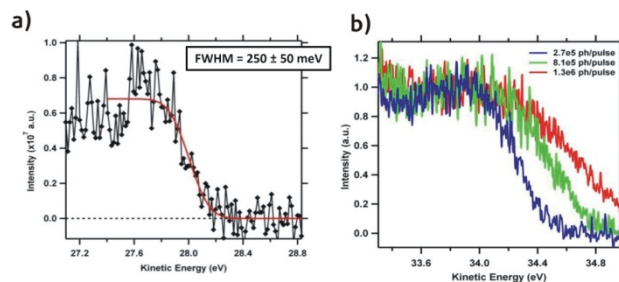


Figure 4: a) Fermi edge measured on TaS₂ with the 17th harmonic of Ar (28 eV). The overall photoemission resolution is 250 meV deduced from the fitting of the edge (red curve). b) Effect of the XUV power on the Fermi edge (broadening and shifting). Above $3 \cdot 10^5$ ph/pulse the space charge effects become non negligible for a $50 \mu\text{m} \times 50 \mu\text{m}$ spot size on the sample.

Carrying out photoemission experiments with a 1 kHz pulsed source, one has to be careful to the photon flux due to space charge effects. Indeed if too many electrons are emitted in the vacuum by a single photon pulse, those electrons will interact by Coulomb repulsion in the region very close to the sample surface. Consequently the spectral information (kinetic energy) from the photoemission process will be altered. Figure 4b shows the Fermi edge of TaS₂ measured with the 21st harmonic at three XUV fluxes (normalized at 33.8 eV kinetic energy). The broadening and shift of the edge is a signature of space charge effects and imposes a flux upper limit of $\sim 3 \cdot 10^5$ ph/pulse for a $50 \mu\text{m} \times 50 \mu\text{m}$ spot size on the sample.

The time resolution of the monochromatized XUV beam was determined by measuring the Kr²⁺ ion yield using an ion time-of-flight spectrometer. The Kr atom is pumped with the 23rd harmonic (35.7 eV) producing Kr⁺ ion in a long-lived excited state. The 800 nm (1.55 eV) probe beam can then ionize the excited ion to Kr²⁺ for positive delay time between the pulses. From the sharp step in the Kr²⁺ yield we extract the XUV pulse duration of 24 fs starting with an IR pulse of 28 fs. Further details about this experiment are presented in this year annual report by W. Bryan *et al.*

Conclusion

The XUV beamline with the photoemission experimental chamber are now available to users. Our first time resolved ARPES measurements are presented in this year annual report by J.C. Petersson *et al.*

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Spin and Angle resolved photoemission with fs laser source: calibration of spin detector

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The experiments presented here were carried out at the Sincrotrone Trieste within the frame of FERMI@Elettra free electron laser project through an international collaboration with the CLF/Artemis project. Here a fs-laser source is combined to the Time-of-Flight-Spin electron analyzer developed at STFC-Daresbury for performing spin, angle and time resolved photoemission spectroscopy experiments on solids.

Introduction

Angle resolved photoemission is a very powerful technique to explore the electronic structure of materials in order to understand their macroscopic properties. Over the past three decades this spectroscopic technique has well matured partly due to the development intense synchrotron radiation source and bright fs-laser source. Measuring the spin as well as the emission angle of the photoelectron is particularly interesting to study the magnetic properties of ferromagnetic systems or systems with large spin-orbit coupling [1-2]. The intrinsic low efficiency of the electron spin detection makes these experiments very challenging. This is why, combining very bright photon source (like table top laser source) with novel electron analyzer (electron time-of-flight) is a real step forward for this type of experiments particularly in term of acquisition time and resolution. We have developed an electron time-of-flight (ToF) spin analyzer where all the electrons are spin and energy- resolved simultaneously regardless of their kinetic energy. Essentially a very high efficiency electron detector (ToF) is combined to a spin detector (mini Mott polarimeter).

The spin polarization P of an electron beam is defined by the asymmetry $P = (n^{\uparrow} - n^{\downarrow}) / (n^{\uparrow} + n^{\downarrow})$, where $n^{\uparrow(\downarrow)}$ is the number of spin up (down) electron. In order to extract the spin polarization from the experimental data the spin sensitivity of the polarimeter, *i.e.* the Sherman function, must be evaluated. Very few precise spin calibrations have been reported so far in the literature because of the difficulty of producing either an electron beam of well known spin polarization or a fully spin polarized electron beam. However an interesting source of highly polarized electron is the Au(111) Surface States (SS) which are free electrons localized at the metal surface. Enhanced spin-orbit coupling (the Rashba-Bychkov effect) is observed due to the high atomic number of gold. In the following work we are using both SS as reference to perform the calibration of the Mott polarimeter.

Laser system and electron analyser

The laser system used is a regenerative amplifier system (RegA9050 from Coherent) providing photon pulses at a

repetition rate of 250 kHz with a pulse energy of 5 μ J and a photon energy of 1.55 eV (800 nm). The 4th harmonic (6.2 eV) of the fundamental beam was obtained by harmonic generation in phase matched BBO crystals. The beam size on the sample was 100 μ m diameter with 5 pJ pulse energy and 160 fs pulse duration.

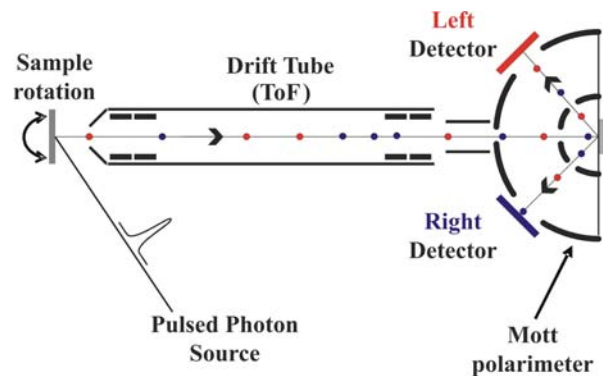


Fig.1 : Top view of the experimental setup in normal emission geometry. The photoelectrons travel through the drift tube up to the Mott polarimeter where they accelerated to 20 kV and the back scattered beam is measured. The arrival time of the electrons at each detector is measured and converted to kinetic energy. The vertical spin polarisation is resolved with the Left-Right detectors of the Mott polarimeter.

In an ultrahigh vacuum (UHV) chamber (10^{-10} mbar) a Au(111) single crystal was mounted on a sample holder with control of the polar (theta, vertical axis) and azimuthal (phi, surface-normal axis) angles. The surface was prepared by Ar ion sputtering and annealing to 400°C. The reconstruction of the surface was monitored using Low Energy Electron Diffraction.

A sketch of the ToF analyser and Mott polarimeter are presented in Fig.1. The ToF analyser is composed of input electron optics, a 25 cm long drift tube and output electron optics which taken together position the Mott polarimeter scattering foil 45 cm from the sample. In the particular case of very low photon energy, all the ToF electrodes are grounded and the electron trajectory in the ToF is considered as ballistic. The angular resolution is then given by the 3 mm diameter entrance hole of the Mott polarimeter and is estimated to be $\pm 0.19^\circ$. All the electrons within this solid angle are accelerated in the Mott polarimeter, hit the targeted gold foil and the back-scattered electrons are measured on four sets of Micro Channel Plate (MCP) detectors. The maximum energy lost in the scattering process is controlled by the Mott polarimeter energy window which was set to 500 eV. The electron arrival time is measured on each detector by a Time-to-Digital-Converter (GPTA mbH) with a time

digitization of 120 ps, the arrival time was then converted to kinetic energy. The overall time resolution of the electronics was 293 ps corresponding to an ultimate energy resolution of 2 meV. The energy resolution of the measurements presented here is evaluated at 25 meV limited by the band width of the laser pulse. The vertical spin polarization of the electrons is deduced from the left-right detectors of the polarimeter. A top-bottom detector pair is available for the horizontal spin polarization (not shown in Fig.1).

Determination of the Mott polarimeter spin sensitivity

At normal emission both spin up and down SS have the same binding energy due to the fact that the momentum of the electrons in the initial state is zero. As the sample is rotated electrons with larger parallel momentum are detected and the fully spin polarized SS are splitted. For a sample angle of -12.2° between the surface normal and the ToF acceptance direction, the averaged EDC (Fig.2-a) obtained from the Mott polarimeter four detectors shows a broad structure just below the Fermi edge corresponding to two peaks. At room temperature the thermal broadening is larger than the energy splitting so to determine the positions and the width of the SS we performed a double Voigt function fitting (solid line). A flat background was subtracted in order to remove the unpolarized contribution from the sp-band. On the averaged EDC both SS have similar total intensity whereas the EDCs measured with the Left (Fig.2-b) and Right (Fig.2-c) detectors show different ratios according to the spin polarization of the photoelectrons.

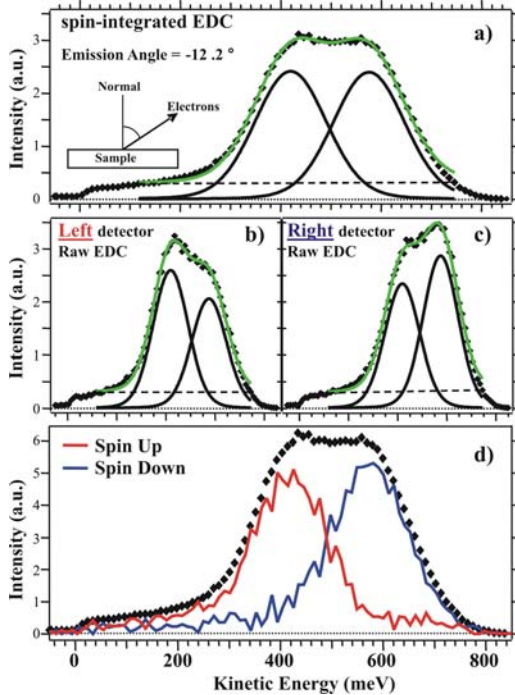


Fig.2 : a) Spin-integrated EDC (diamonds) obtained by averaging the four detectors of the Mott polarimeter. The surface states energy positions and widths are extracted using a double Voigt fitting function (black curves) assuming a flat background. b)c) Left-Right detector EDC fitted with the parameters from a). Only the amplitude of the surface states is left as a free parameter. d) Spin-resolved EDC (red and blue curves) obtained from the EDC spectra of panel b) and c).

The four total intensities of the SS measured in Left-Right detectors are deduced from the Voigt fitting of the raw EDCs Fig.2-b-c. Using a symmetric relation between those quantities

one can extract the experimental asymmetry and the effective Sherman function of the Mott polarimeter. In the present case we obtained $A_{\text{exp}} = 2.6 \pm 0.2\%$ and $S_{\text{eff}} = 10.8 \pm 0.7\%$ for an accelerating voltage of 20kV in the Mott polarimeter. Finally the spin-resolved EDC (Fig.2-d) are calculated with the relation $I_{\uparrow(\downarrow)}(E) = I(E) \cdot (1 \pm P(E))$ where $I(E)$ is the average spin integrated spectrum (Fig.2-a). Fig.2-d shows that both SS are clearly resolved with a constant sp-band background. With a photoelectron trajectory calculation we obtain a real emission angle of 19.1° (20.2°) for SS1 (SS2) of Fig.2-d which correspond to the momentum $k_{\text{SS1}} = 0.126 \text{ \AA}^{-1}$ ($k_{\text{SS2}} = 0.114 \text{ \AA}^{-1}$). Those results are in very good agreement with ARPES measurements performed at higher photon energy [3].

Variation of the Mott polarimeter high voltage

The scattering efficiency of the Mott polarimeter is defined by the ratio I/I_0 where I is the current measured in the detectors and I_0 is the current injected in the polarimeter. The spin efficiency of the Mott polarimeter depends strongly on the accelerating voltage V_{Mott} applied on the target gold foil.

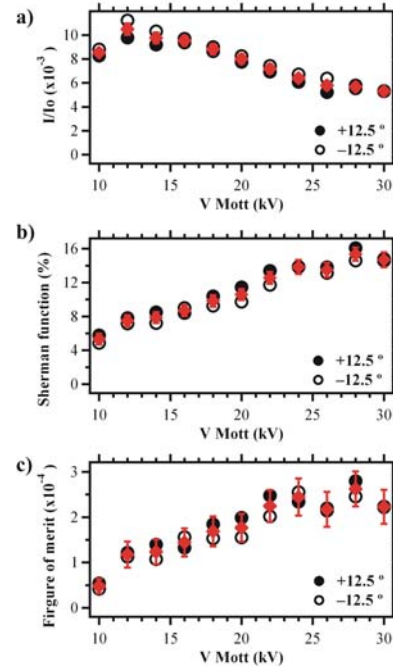


Fig.3 : a) The scattering efficiency I/I_0 decreases with the Mott polarimeter high voltage. The measurements were performed at $+12.5^\circ$ (circle) and -12.5° (solid circle) from normal emission. Red diamonds are the average intensity. b) The value of the spin selectivity of the Mott polarimeter (Sherman function) increases up to 15% with the accelerating voltage. c) As the Mott high voltage increases, the Figure of merit increases to reach a plateau at 25kV.

In Fig.3 we present the variation of both the ratio I/I_0 (Fig.3-a circles) and the S_{eff} (Fig.3-b circles) measured in two photoemission directions ($\pm 12.5^\circ$ from the surface normal) and for various V_{Mott} values between 10kV and 30kV. The average values are deduced (Fig.3 diamonds) and the error bars are calculated from the fitting parameter errors for a confidence interval of 95%. The dispersion of the measurements for both sample angles is within the error bars. In the voltage region 20-30 kV the effective Sherman function is measured with an

overall error of $\pm 6\%$ which is consistent with the best results reported in the literature. Thus setting our apparatus as a reliable and high performance system for spin-resolved ARPES. Between 10-30 kV accelerating potential the scattered intensity decreases by $\sim 50\%$ whereas the Sherman function increases by $\sim 300\%$. This large increase of the Sherman function is consistent with the increase in the gradient of Coulomb potential (and so the spin-orbit interaction) as the incident electrons travel closer to the Au nucleus.

Assuming a merely statistical error for the measurements ($\Delta I = \sqrt{I}$) the spin polarization error is $\Delta P = 1/\sqrt{IS^2}$ where IS^2 is the figure of merit (FOM) of the Mott polarimeter. Fig.3-c shows the FOM calculated from the results of Fig.3-a-b. The

FOM increases with V_{Mott} to reach a plateau at ~ 25 kV, the optimal voltage to operate the ToF-Spin analyzer.

Conclusion

We have performed spin and angle-resolved photoemission on Au(111) at very low photon energy. The good efficiency of the electron ToF-Spin analyser allowed us to work with high angular and energy resolution while still maintaining a short acquisition time (~ 10 min). The spin-orbit coupling of the SS was spin-resolved and analysed as a fully spin polarized electron source. The effective spin sensitivity of the Mott polarimeter was extracted from the raw measurements for various Mott voltages up to 30kV. The figure of merit shows that the optimal condition to operate the Mott polarimeter is ~ 25 kV [4].

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Few-cycle carrier-envelope-phase controlled laser pulses for time resolved science at the Artemis facility

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Introduction

When the duration of a laser pulse reaches the few-cycle level, the phase of the laser electric field relative to the pulse envelope (the carrier-envelope phase, CEP) can have a significant effect on the physics of the interaction. Examples of this are the generation of isolated attosecond pulses [1] and the control of electron localisation during molecular dissociation [2], both of which require few-cycle pulses with stable carrier-envelope phase.

The Artemis facility [3, 4] for ultrafast XUV science is constructed around a high power CEP stabilised laser system with an output of 780 nm, 30 fs, 14mJ/pulse, 1kHz. Part of the energy can be split off to generate few-cycle CEP-stabilised pulses at 780nm and tuneable pulses from the UV to the mid-IR. These can be flexibly configured to produce femtosecond XUV pulses through high harmonic generation (HHG), or used as pump and probe pulses. We have recently made improvements to the few-cycle beamline to reduce the pulselength to 7 fs and measured the long-term performance of the CEP stabilisation.

Carrier-envelope phase control

The laser is CEP stabilised using commercial f-to-2f interferometers to measure the CEP drift and feedback electronics. A fast CEP loop feeds back to the end mirror in the oscillator to ensure that every fourth pulse of the 80 MHz repetition rate output of the oscillator is identical. The slow CEP loop acts on a compressor grating to ensure stability of the amplified pulses. We are able to lock the CEP of the laser at full power to 310mrad rms (measured with 10 ms integration time) for over an hour (Figure 1).

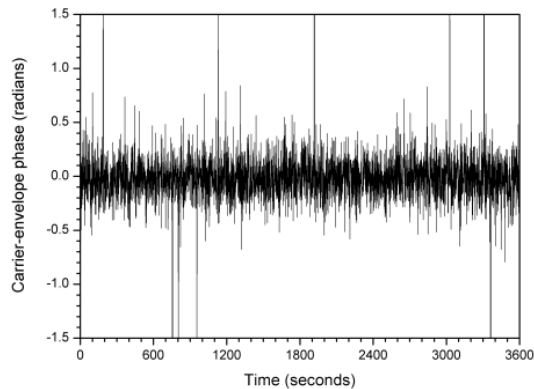


Figure 1. Measured carrier-envelope-phase stability of the Artemis laser system: (a) 310 mrad rms noise over one hour with 13.5 mJ/pulse.

When running the laser with 2mJ/pulse (for compression in the hollow-fibre) we are able to control the phase over a period of five hours (Figure 2) with rms stability of 325 mrad. In this plot, the phase was intentionally ramped so we could acquire data.

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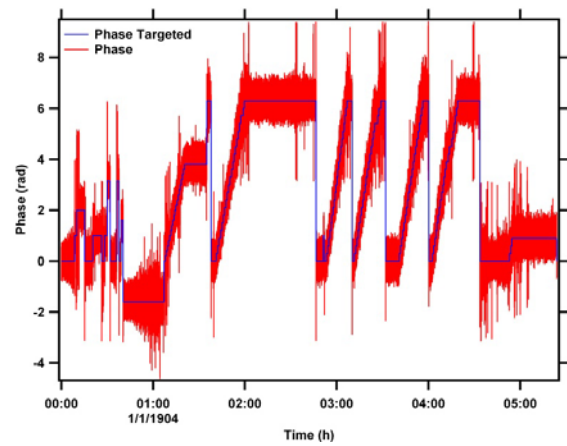


Figure 2. CEP control over five hours with 2mJ/pulse and 325 mrad rms stability. Blue line: programmed target phase. Red line: measured phase.

Few-cycle pulses

Few-cycle laser pulses are produced by splitting off 1 mJ of the laser output and spectrally broadening it in a gas-filled hollow fibre, as previously used in Astra-TA1 [5]. The laser pulses are then recompressed using chirped mirrors. The spectral phase of the few-cycle pulses is measured with SHG-FROG. With 2.2 bar neon pressure in the fibre we obtain 7 fs, 0.5 mJ pulses (Figure 3). This is a substantial improvement on the performance we achieved in Astra-TA1 (which was typically ~ 10 fs). This is due to a combination of changes in focusing into the fibre, improvements in alignment diagnostics, better chirped mirrors and increased bandwidth in the FROG through using a thinner doubling crystal (10 μ m BBO).

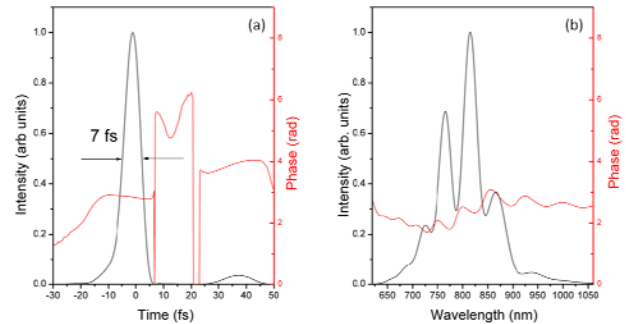


Figure 3. FROG measurement of 7fs, 0.5mJ pulses produced in the hollow fibre system: (a) temporal intensity and phase, (b) spectral intensity and phase. The FROG retrieval error was <0.006.

Conclusions

We have characterized the Artemis few-cycle, carrier-envelope-phase controlled beamline. We measured 7 fs, 0.5 mJ pulses at 1 kHz and have achieved CEP control over a period of 5 hours with rms phase stability of 325 mrad. We have planned further

improvements to the diagnostics and data acquisition with the aim of reducing the pulselength and rms CEP values still further.

Acknowledgements

We thank our technical staff, especially Steve Hook and Toby Strange.

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Vibration in Gemini and Engineering modifications (interim report).

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Introduction

Astra Gemini is a dual beam facility with each beam delivering 0.5PW energy in 30fs every 60 seconds. Gemini is housed in building R7, the seed beam comes from Astra and is split, amplified and compressed above the Target Area then delivered through the bunker roof (300mm thick concrete) into the Target Area. In the Target Area each beam propagates off a number of mirrors to be focused to a 2-20 micron spot at the interaction point where they interact with extremely delicate targets which are vaporised. Experiments can utilise both beams interacting together and this requires a highly stable and synchronised system in both space and time.

Vibration was observed within the target area that affected both the internal and external laser beam pointing stability. The observed motion of the focal spot depended on the nature of the focussing geometry but often created a very stable elliptical or figure of eight movement of the spot. This was observed to disappear completely during maintenance periods in ISIS when the main magnet power supply was switched off. The motion produced by the vibration was observed to be about 6-10 spot diameters. This degree of motion is detrimental for most experiments and catastrophic for any dual beam spatially synchronised operation.

ISIS has a number of different systems used to generate the stable 50Hz power required to run. A generator is used predominately with a backup system employing a UPS. To determine the exact source observations were carried out with the generator running and the UPS running. No obvious difference was observed ruling the generator out as the source. Physical inspection of the choke and area round the choke identified a strong vibration which propagated into R6. The choke is one of the main power components for the magnets that form the synchrotron.

Figure 1 shows a simplified site plan identifying the relationship between the Gemini Target Area and the choke. The choke is just outside building R6 and tunnel 7 joins building R6 to building R7. Figure 2 shows a plan view from Google Maps identifying the same buildings and the area of the tunnel. Figure 3 shows the target chamber mounted directly over the trench leading directly to tunnel 7. Both the breadboard support frame and the chamber support frame straddle the trench. The mounting positions directly over the trench are unused and are clear of the trench covers. Tunnel 7 is a concrete structure approx 2.5m high and 2m wide inside and is used for services infrastructure. The whole of the site is built on a chalk bed and recent tests on the chalk 100m away from building R7 suggest the chalk is nearly as dense as structural concrete.

The vibration was observed on the monitors connected to the alignment microscopes which are used to align the beam to the target.

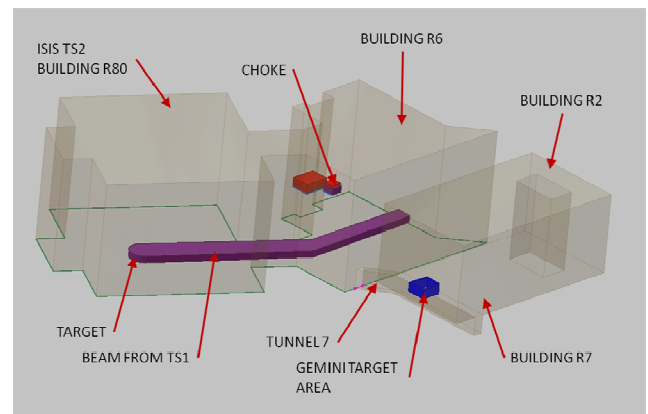


Figure 1-Relationship between the choke and target area

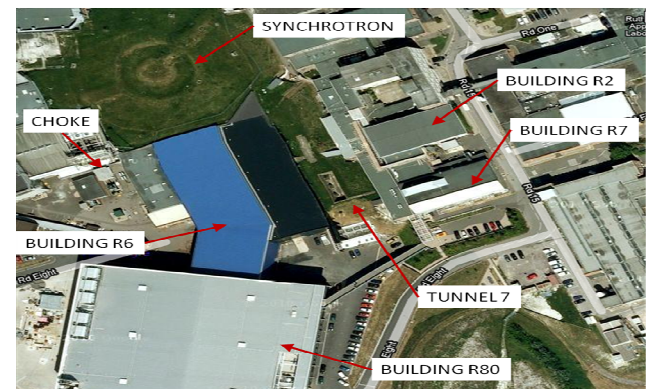


Figure 2-Plan view from Google Maps of the area.

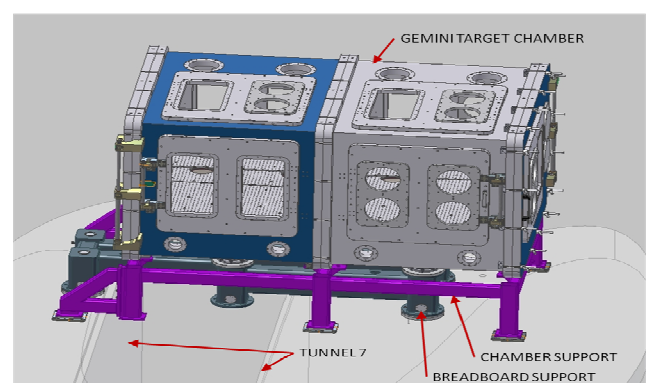


Figure 3-Relationship between the target chamber and tunnel 7

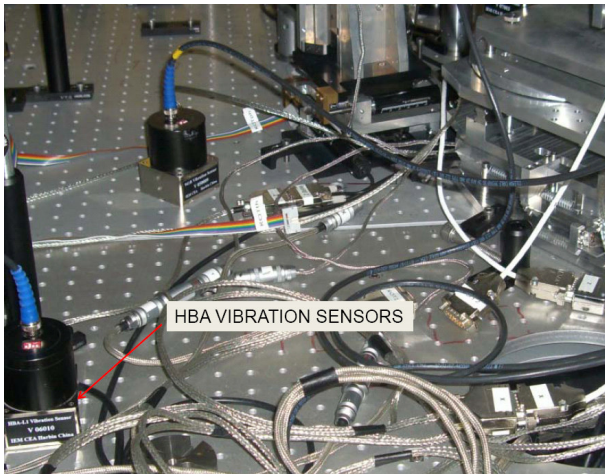


Figure 4-Vibration measurement being carried out inside the Gemini Interaction Chamber

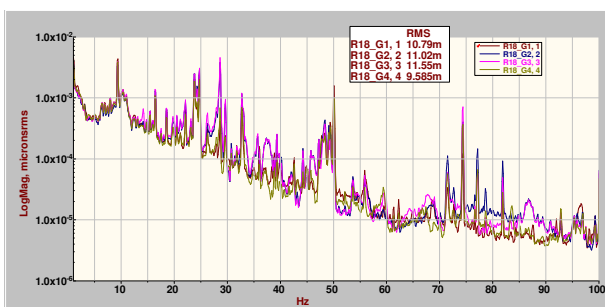


Figure 5-Frequency spectrum measured in the Target Area with ISIS off.

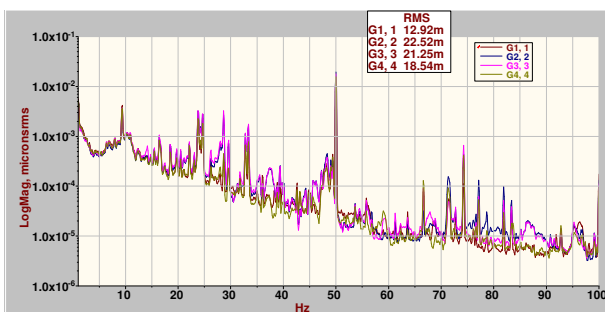


Figure 6-Frequency Spectrum measured in the Target Area with ISIS on.

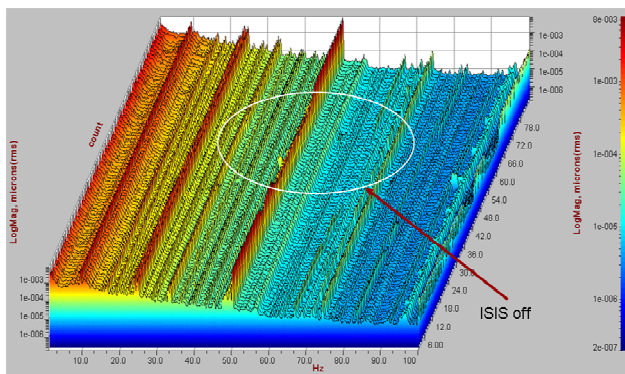


Figure 7-Spectrum plot of one sensor showing ISIS switch off and the corresponding change.

Vibration Survey

A vibration survey was carried out in collaboration with Houch-Eng Huang in Diamond. Figure 4 shows the HBA sensors measuring positions on the breadboard. These sensors are used routinely in Diamond around the ring and the sensor design is optimised for sensitivity to ground borne vibration down to 0.5Hz.

Figures 5 & 6 show plots of the spectrum with ISIS on and ISIS off. There is a clear increase in the spike at 50Hz and a number of other spikes that appear around 30Hz and 70Hz. Figure 7 shows a direct comparison of displacement against time for one sensor with and without ISIS. The signal propagated is only visible in the vertical plane.

Figure 8 shows the spot movement of a He Ne laser propagated off a number of mirrors both in the target area and in the interaction chamber itself. The spot position was recorded over several milliseconds with a lateral effect position sensitive detector. A Fourier analysis reveals a strong 50Hz component as identified in the other vibration measurements. It should be noted that the movement is much more pronounced in the horizontal direction but could still be caused by vertical ground borne vibration due to the complicated beam path involving periscopes. Further investigation is underway.

Considered methods to overcome the problem.

1. The laser could be synchronised with the ISIS 50Hz power supply making the laser fire at the same exact time in the cycle allowing alignment. The CW beam would additionally be pulsed with the 50Hz so that it also appeared to be stationary.
 2. Investigate the TA hardware to identify items with a natural frequency at 50Hz.
 3. Isolate the chamber from the source with Sorbothane, a material known for its damping characteristics.
 4. Isolate the source of the ground borne vibration.
 5. Break the main path to the chamber with a trench.
 6. Stiffen all the structure so that it performs as a single entity.
- (1) The effort and cost required to synchronise with ISIS was small and this was the first step.
- (2) Initial tests on the chamber components were carried out using a SKF RT440 looking for a significant increase in 50Hz amplitude throughout the structures. Initial testing suggested the signal was amplified through the breadboard structure and further amplified through the parabola mounts.
- (3) Isolating the breadboard in the chamber was ruled out. The ground borne vibration was measured throughout the bunker. With the whole bunker being used to support hardware in the laser chain it is expected that isolating one item would only move the problem to manifest itself somewhere else. Additionally major work on the breadboard would require a complete strip of the area which would see a shutdown of >6 months. This would have a serious impact on operations and was deemed unfeasible.
- (4) The source is significant hardware at the heart of ISIS. Isolating this is not feasible. It is currently identified as an area of single point failure as is no longer commercially available. A replacement unit is in construction in building R6 forming 8-10 smaller units. Commissioning is planned later this year but the impact is difficult to determine.
- (5) Breaking the path with a trench is not feasible due to there being no practical location for the break. The cost and impact to operations caused by the work would be significant.
- (6) Stiffening the structure is possible and feasible.

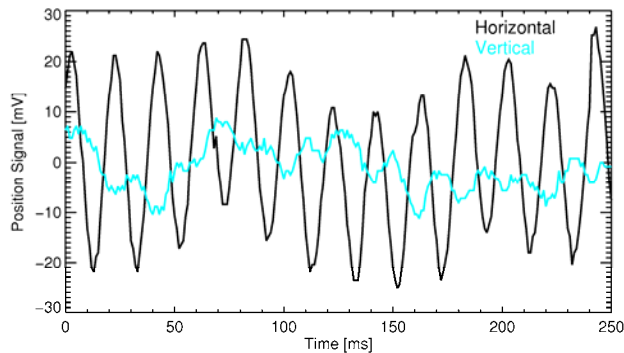


Figure 8-Spot movement of a He Ne laser propagated in the Target Area.

Components with a natural frequency of 50Hz

The Parabola mounts were both found to amplify the 50Hz signal and were dispatched to TUV for full spectrum testing. Figure 9 shows the mount on test on a shaker table. The mount is designed to align the parabola and is a high accuracy five stage translation and rotation stage utilising high precision commercially available motor micrometers with zero backlash.

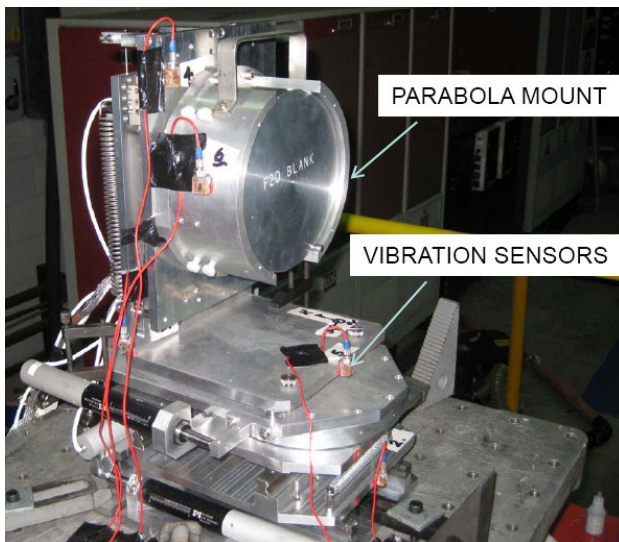


Figure 9-Parabola mount under test at TUV.

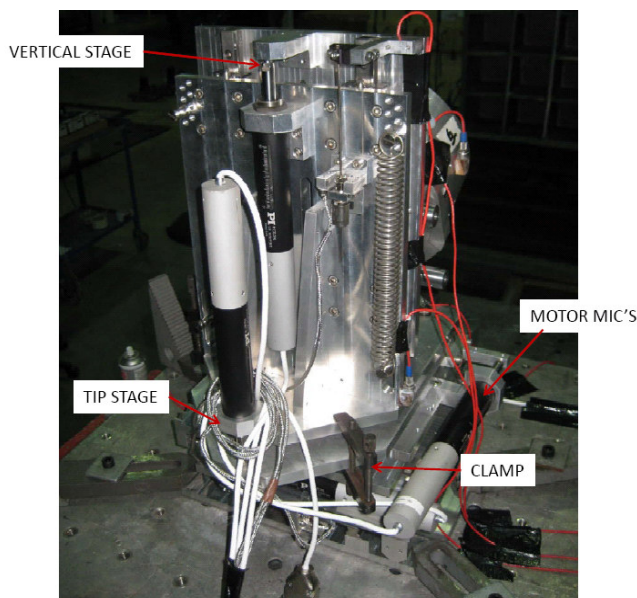


Figure 10-Tip and Vertical stages shown to be sensitive.

Figure 10 shows the back of the mount where the vertical and tip stages are located. The clamp shown was used to methodically stiffen the mount allowing re-runs to identify the sensitivity to vertical vibration in each stage. The mount has two different configurations depending on its location within the Interaction Chamber. Both were tested with sweeps up to 105Hz using the lowest input "g" level the machine could register a signal. Figure 11 shows the control channel at 0.03g.

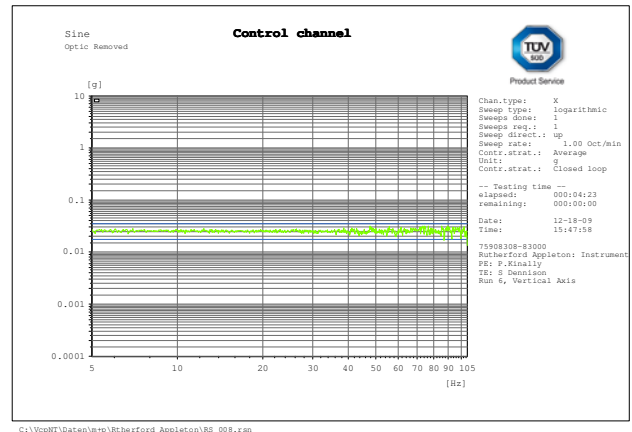


Figure 11-Shaker rig control channel at 0.03g

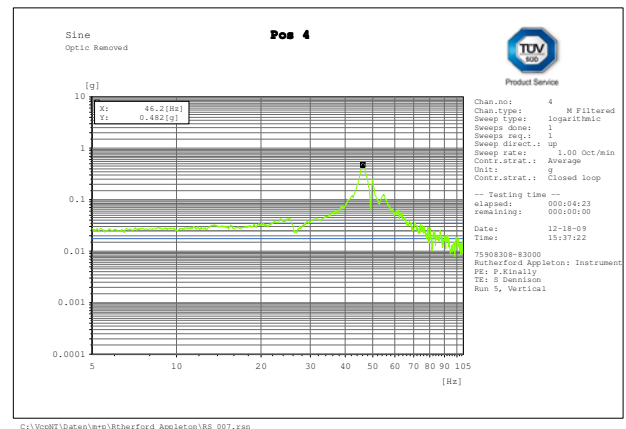


Figure 12-Shaker rig scan identifying problems in Tip Stage.

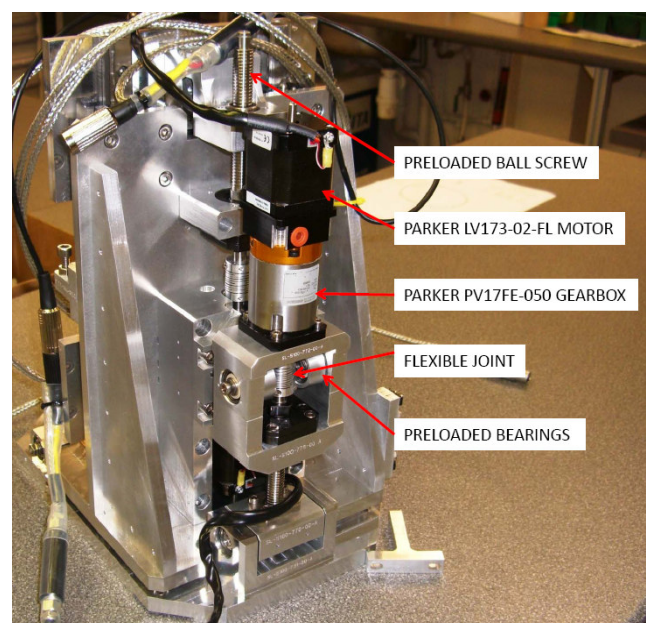


Figure 13-New parabola mount tip and vertical stages.

Figure 12 shows the tip stage having a natural frequency at around 46Hz and 50Hz. The vertical stage had a similar graph at 50Hz.

The original design intent was to manufacture an extremely compact mount capable of being positioned side by side to allow both 0.5PW beams to be focussed on target using commercially available hardware with zero backlash. To overcome the 50Hz natural frequency it was obvious a re-design stiffening the tip and vertical stages was necessary. Figure 13 shows the new design implemented. The two Pi motor micrometers have been replaced by high precision preloaded ball screws from THK. The Parker motor and gearbox are similar to that used in other areas across the facility without problem. The preloaded bearings were purchased from Carter bearings. With preloaded bearings and preloaded ball screws it is expected that the stiffness in the vertical direction will be significantly increased.

Stiffen all the breadboard structures

The design intent for the breadboard in the Interaction Chamber was for it to be populated and aligned with optics offline allowing a faster turnaround of experiments and more experimental output. The breadboards were split up into four separate pieces. This is shown in figure 14. The four separate breadboards were bolted to two separate intermediate frames then to the main sub-frame. Initial analysis suggested that this structure amplified the 50Hz signal and therefore had a corresponding natural frequency.

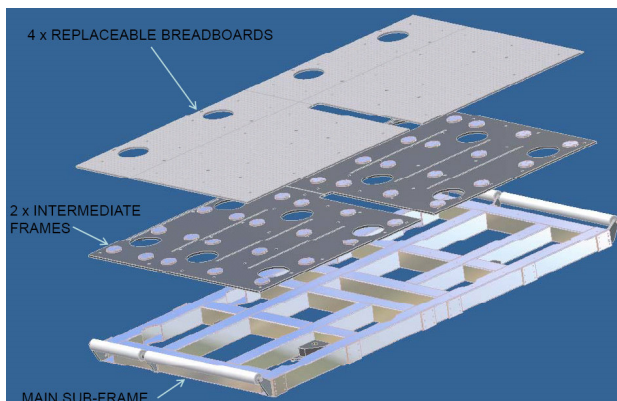


Figure 14-Initial breadboard construction

Time was short for a detailed investigation and the decision was made to consolidate all the components into one single structure shown in figure 15. The support legs were also revised to improve the installation procedure. The 7075 aluminium had a longer lead time over cast tooling plate but 7075 offers better scratch resistance over cast tooling plate and offers longer wear for the threads. 7075 aluminium suffers from its notch sensitivity leading to crack propagation however its notch sensitivity would not be a weakness in this design.

Figure 16 shows the long focal length parabola chamber which houses a parabola mount and the F20 optic. This assembly was stiffened by increasing the thickness of the floor plate, stiffening the support frame and introducing bellows to reduce the bending caused by the deflection in the main chamber during pump down.

Further stiffening was carried out by casting the support structures to the floor using Fosroc Conbextra EPR for the larger structure and Fosroc Conbextra EPLV for the feet.

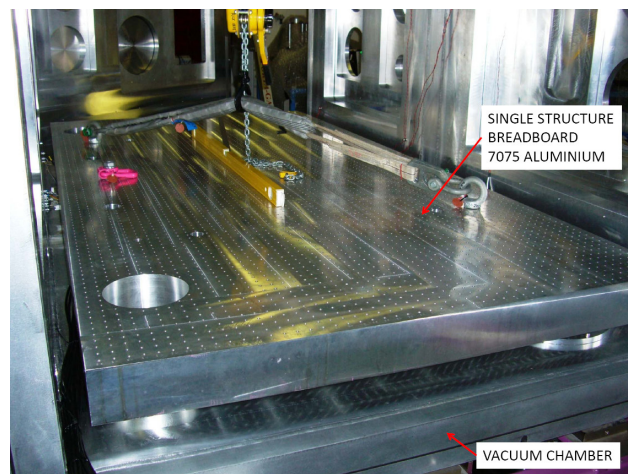


Figure 15-Revised single structure breadboard

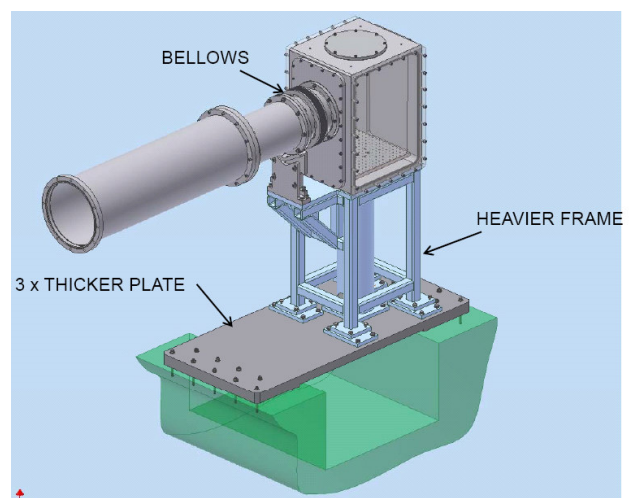


Figure 16-revised structure for the long focal length parabola

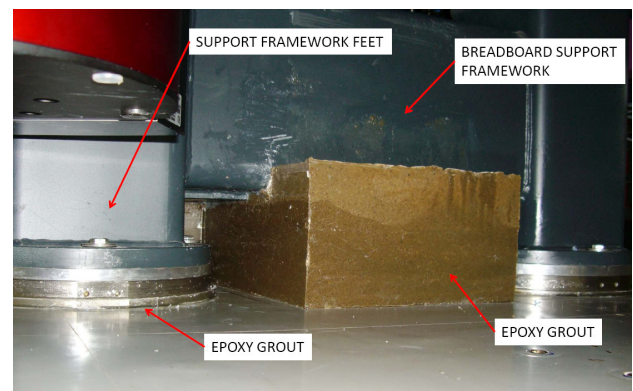


Figure 17-The breadboard support structure is cast to the floor

The EBR variant is a loaded two part epoxy compound and McNaughton Bailey Ltd were employed to weir up the structure and pour the material over a weekend. The trench limited the length of the pour which extends from the east side shown in figure 17 to the edge of the trench. Stopping at the trench ensures continued access for services. The EPLV material is a very low viscosity two part epoxy which was flooded in around the feet ensuring a good connection to the concrete substrate below.

Analysis of the revised structure

Figure 18 shows the location of the vibration sensors for the plot on figure 19. Sensor 4 is assumed to be the reference as the structure is now bonded to the floor. Sensors 1-3 are therefore measuring any amplification of the signal through the structure. Although the breadboard structure was replaced it was not possible to make any structural changes to the four support legs without dismantling the chamber. Figure 19 shows the spectrum with ISIS switched on. There is a marked reduction in the background over the initial scans. Sensors 1 & 3 produce an identical trace and show only a slight increase over the reference. Sensor 2 shows a further increase that could suggest that the trench is the main conduit for the vibration or that further stiffening of the structure above the trench could provide further gains.

Conclusions

Replacing the breadboard with a single structure, grouting the breadboard support to the ground and reinforcing the long focal length parabola have all significantly reduced the background noise, but as expected the main peaks from ISIS are unaffected.

The single breadboard eliminates the previous local problems where components spanned two sections of the breadboard. Initial experimental operations on the new structure suggest the structural changes are positive. The changes to the parabola mounts are expected to have a significant improvement again. The area is operational preventing the new parabola mounts being installed and they are on standby should there be problems with the current mounts prevent them from being tested on the shaker table.

All areas on site see ground borne vibration from ISIS, Diamond and traffic on the A34. In this particular area we had structure that was sensitive to 50Hz. Following the design philosophy included herein should prevent the need for expensive computer analysis on all future designs.

This is an interim report pending the results of the vibration sweep of the new parabola mount design and final testing in the vacuum chamber. Final conclusions will be in next year's report.

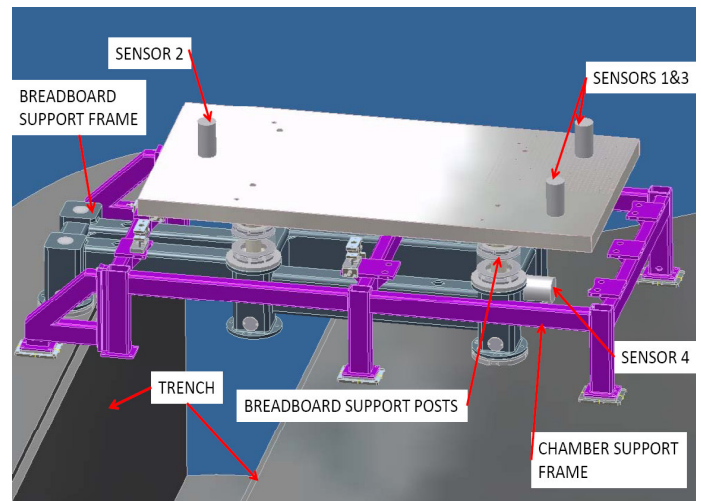


Figure 18-Showing the new structure and the location of the four sensors

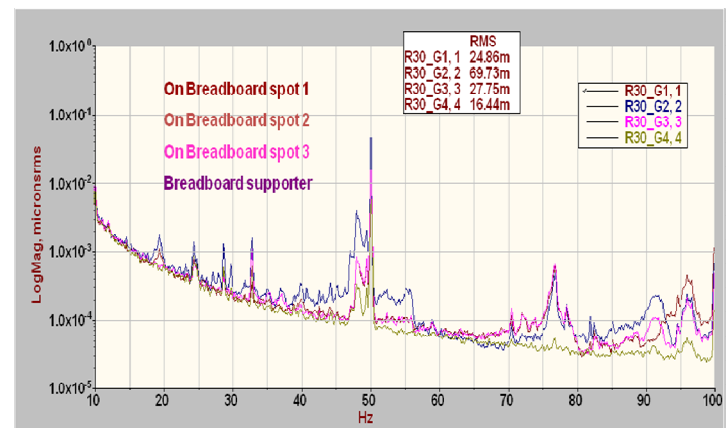


Figure 19-Measurements taken of new breadboard structure

Temporal and Spatial Overlap Monitoring of the Dual-Beam Layout in Astra-Gemini TA3

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Introduction

The Astra-Gemini target area has been operational for user experiments since 2008 in a single beam configuration. The laser system is now up and running for dual beam experiments. In order to meet the requirements for users with small focal spot sizes ($\sim$ few microns) and femtosecond pulse durations it is now critical for the outcome of experiments that the spatial and temporal overlap of the two beams at the target position. Here we report on two newly implemented diagnostics in the target area to monitor the overlap (both spatially and temporally) of the two beams.

Astra-Gemini TA3

The current experimental set-up is to have the two beams of the Gemini system focused onto target with the $f/20$ parabola on the south beam, focusing to a spot of $\sim 30 \mu\text{m}$ and the north beam focused with an $f/2$ parabola, giving a spot of $\sim 2 \mu\text{m}$. The pulses are compressed to pulse durations of $\sim 40 \text{ fs}$. With the implementation of the first dual beam experiment using Astra-Gemini, the size of the focal spots and the ultra-short pulse duration of the system means that the need to have shot-to-shot measurements of the spatial and temporal overlap of the two beams is of extreme importance. These extra diagnostics are added into the existing TA3 beam diagnostics which use the leakage through the rear of one dielectric mirror in each of the beam lines. A schematic diagram of existing and planned beam diagnostics is shown in Fig. 1. For each experimental arrangement, once zero delay between the main beams has been set up, the delay in the south diagnostic line can be adjusted to reset the temporal overlap monitor.

Beam Overlap Diagnostic

For the first overlap diagnostic, two leakage beams are combined using a dielectric beamsplitter inside the target chamber after the minimum possible number of optics. Because very little energy is needed we pick off a $\sim 1 \text{ cm}$ beam from the unfocused mirror leakage. The combined beam is directed to a system above the target chamber built with caged opto-mechanics to increase stability, shown in Fig. 2(a). The beam is focused using an $f/20$ lens and split onto a pre-focus near-field camera and a far-field camera with a microscope objective. A slight adjustment on one of the diagnostic beams then produces two closely spaced but distinct focal spots and interference fringes in the near field.

Once the beams are combined, vibrations in the common optics are not important since we need to monitor only the relative positions of the two focal spots. It will be necessary to check whether instabilities in the leakage beam focal positions correspond exactly to those in the main beam focal spots.

The far-field camera will measure the shot-to-shot spatial stability of the two beams. Software will be developed to monitor the positions of the spots and later to provide feedback to a pointing mirror to allow for automatic correction of beam drift.

The visibility of a near-field interference pattern will be an accurate indicator of a temporal overlap between the two beams

of less than the pulse envelope duration. It will also be sensitive to jitter in the spatial alignment since this will alter the fringe pattern.

Cross-correlator diagnostic

The second method to monitor the temporal overlap is to measure a cross-correlation signal between the two leakage beams (Fig 2(b)). This requires an energy $\sim 1 \text{ mJ}$ so we down-collimate the leakage beams using a telescope with magnification ~ 15 (the same focusing lens as used for the main beam equivalent plane monitor). The beams are then split and overlapped in two separate BBO crystals to generate two cross-correlation traces, imaged without magnification to CCD cameras. At the zero-delay position this gives a centred vertical stripe. Any change in delay between the beams moves this stripe, the direction depending on which beam comes earlier.

One of the correlators has a ‘tight’ crossing angle which provides a time-window of $\sim 6 \text{ ps}$. A shift in the delay stage of $10 \mu\text{m}$, corresponding to a delay $< 100 \text{ fs}$, is clearly detectable on this camera. The second arrangement has a wider time window to measure delays of $> 6 \text{ ps}$, lying outside the window of the ‘accurate’ timing correlator. Delays $> 50 \text{ ps}$ can be measured using a fast photodiode (35 ps rise time).

We intend to write software to capture the central position of the correlation trace on each shot providing a shot-to-shot measurement of temporal stability. This could also be incorporated into a feedback system to the delay stage in LA3 to correct for timing drift during the day.

Conclusions

The beginning of dual beam operations on Astra-Gemini required additional beam diagnostic to monitor beam overlap. We have started to implement such systems and will start to operate them on full power shots in the next few months. It should be possible to use these diagnostics to implement a feedback system to correct for beam drifts.

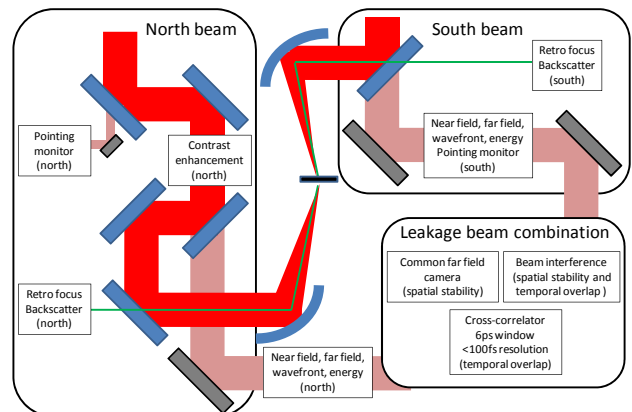


Figure 1: Schematic diagram of the beam arrangements in Astra-Gemini TA3. Red indicates the main beamlines and pink indicates leakage beams.

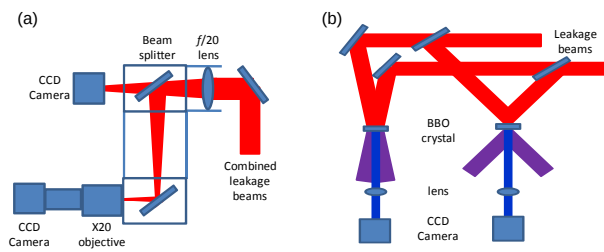


Figure 2: Schematic diagram of (a) the combined beam near and far-field diagnostic and (b) the double cross-correlator arrangement.

Upgrade of Astra Amplifier 3 and the Astra interlock system

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Introduction

The multi-terawatt Amplifier 3 of the Astra facility became operational in 1999¹, and has been in more or less continuous operation ever since. The development of Astra Gemini, with its requirement for better-quality input pulses and higher energy, was the driving force behind changes to Amp 3 made in 2006². These changes included a two-level layout for beam expansion. Although this was undesirable for reasons of stability and accessibility, it was necessary because the large footprint of the Macholite pump laser severely limited space on the table. In the past year that laser has been replaced with four Quanta-Ray lasers which are positioned to one side of the amplifier table, releasing a large amount of space on the table itself. This change provided an opportunity to redesign the amplifier layout and return to a single level. Here we describe the changes made to the amplifier and its infrastructure and present results demonstrating its performance.

New pump lasers and cooling system

The Continuum “Macholite” laser was a customized two-beam Nd:YAG laser producing up to 2.5 Joules of second harmonic light per beam at 10 Hz. In recent years this laser had become increasingly unreliable, being responsible in its final year for nearly half of the lost time on Astra. The new Quanta-Ray lasers each produce a single beam of 1.4 Joules at 532 nm at 10 Hz (1.3 Joules when injection-seeded), so the total available pump energy is 5.2 Joules, slightly higher than before. Another Quanta-Ray laser has been used for many years in Astra to pump the second amplifier, and has proved the most reliable of all the commercial lasers used in the facility.

The cooling water for the Macholite was supplied by a complex system with three circuits, which was hard to access and also prone to faults. To cool the new lasers a completely new cooling system has been installed, with a new external chiller and properly-designed pipework that includes a filter, flow-meter and temperature gauge for each laser. The distribution pipework is mounted on an extension of the gantry that runs the length of the Astra laser area, as are the power sockets for the four lasers.

The lasers themselves stand on breadboards mounted on custom frameworks alongside the amplifier table. Each breadboard has two laser heads side by side, with their power supplies beneath them. The space between the frames is large enough for one power supply to be pulled out for maintenance when required, and the same space allows the laser operators access to the amplifier table for alignment.

Amplifier design changes

In redesigning the amplifier, we took the opportunity to change several of the less satisfactory aspects of the original design. The spatial filters between passes 1 & 2 and 3 & 4 were replaced with a better version based upon the successful design used in the Gemini amplifiers. In this the pinhole is mounted in a fixed position on a base plate that is clamped to the table, and a removable cover with a top viewing window is fitted over it. The beam tubes of the spatial filter have independent supports.

The beam is aligned through the lenses and windows of the spatial filter and referenced to the kinematic crosswires at either end. The pinhole is then positioned so it is centred on the beam (at low power), and finally the cover is placed over the baseplate and the beam tubes joined to the sides. The same achromatic lenses were used as in the old amplifier; these are mounted on slides so the focusing can be adjusted once the spatial filter is under vacuum.

Another change from the previous layout was to allow more space around the TiS crystal holder. It was anticipated that at some stage a cryogenically-cooled crystal might be used in this amplifier, which would need more space, so the beam angles through the crystal were increased slightly, and the spatial filters are a few centimetres further from the crystal mount.

With the pump lasers mounted to one side of the amplifier table there is enough space available to position the entire amplifier so it lies on a single table, rather than straddling across two tables as in the previous design. This allowed the heights of all the components to be preset to the beam height of 110 mm, which ensures the beams are horizontal everywhere.

Pump beam distribution

As there are now four pump beams rather than two, the design of the pump beam expansion and distribution system had to be changed significantly from the previous arrangement. The beam expansion factor from 12 mm to 18 mm is the same as before, but space constraints meant that the focal lengths of the expander lenses had to be shorter, and it was no longer possible to use nitrogen-filled pipes to enclose the focal regions as there would always be breakdown at the foci. We decided to use vacuum pipes of large enough diameter that two beams could be expanded in each pipe side by side. Two of the beams have further to travel, so their expansion optics are of longer focal length. The optics of all four beams are designed to image-relay a plane near the output of the laser onto the crystal, to ensure the illumination of the pumped region is as uniform as possible.

In practice, the windows of these expander tubes caused significant problems. Despite the use of an oil-free pump in the vacuum system, the antireflection coatings on the insides of the windows gradually became contaminated, which led to the coating degrading and eventually being burned off the substrate. The resulting loss of energy compromised the performance of the amplifier until the window was rotated to a new position. At the time of writing, new window assemblies for uncoated Brewster-angle windows are being manufactured, and it is expected that these will resolve the problem.

As in Gemini, the pump beams are on a higher level than the seed beam, 150 mm rather than 110 mm, so pass through the crystal travelling slightly downwards. The transmitted beams are collected on four (plane) recycling mirrors, which retro-reflect them back to the crystal. There is a significant benefit to pump recycling, as the crystals used in this amplifier typically transmit about 7% of the pump light, which would otherwise be wasted. A similar scheme is used in the Gemini amplifiers.

Amplifier layout

The layout of the amplifier and new pump lasers is shown in Figure 1. All the design drawings were made using a 3-D CAD program to ensure none of the hardware components obstructed any of the beams. In laying out the components, a full-size printout of the base layout was fixed to the table, and cut-outs made for the mount bases and other items to allow accurate positioning. This procedure saved a great deal of time, as the design ensured there were no unresolved alignment issues. With the optics positioned according to the drawings, it was easy to align the beam through the amplifier, with only minimal changes to component positions. In the figure, the four pump beams are shown in red, yellow, green and blue, and the 800 nm seed beam in purple. To avoid cluttering the diagram, only representative components are labelled.

As before, the beam makes four passes through the TiS crystal, with a vacuum spatial filter between the first and second passes and another between the third and fourth. After the final pass it is expanded to 31 mm diameter, as required for propagation to Gemini. Energy controls in the form of a waveplate/polarizer combination and slide-in attenuator mirrors, plus a fast shutter, allow the users to set the shot energy and fire shots on demand, or to use the 10 Hz beam at low or medium energy.

but does not give any gain. Changing the trigger delays on the master box allows the timings of all the lasers to be adjusted together, and the separate delays on the lamp and Q-switch boxes allow the timings of each laser to be optimized individually.

Automatic alignment

The automatic alignment system used in the first two amplifiers has not yet been extended to Amplifier 3. However, the fast pointing control system³⁾ has been reinstalled, as this improves the stability of the beam when it enters the Gemini laser area. It consists of a 2-inch mirror mount with three piezoelectric actuators (of which only two are used), and a far-field imaging system. The piezo-mounted mirror is the last optic before the final pass of the amplifier, and the far-field monitor receives the leakage beam transmitted through the next mirror in the chain, where a lens and a microscope objective focus the beam onto a fixed camera. There are no reflections between the piezo mirror and the camera, so the setup provides an absolute pointing reference. When active, the control software drives the piezo mirror to keep the far-field centred on the reference position, except when thermal changes force the piezo voltage out of range, at which point a manual correction of the mirror becomes necessary.

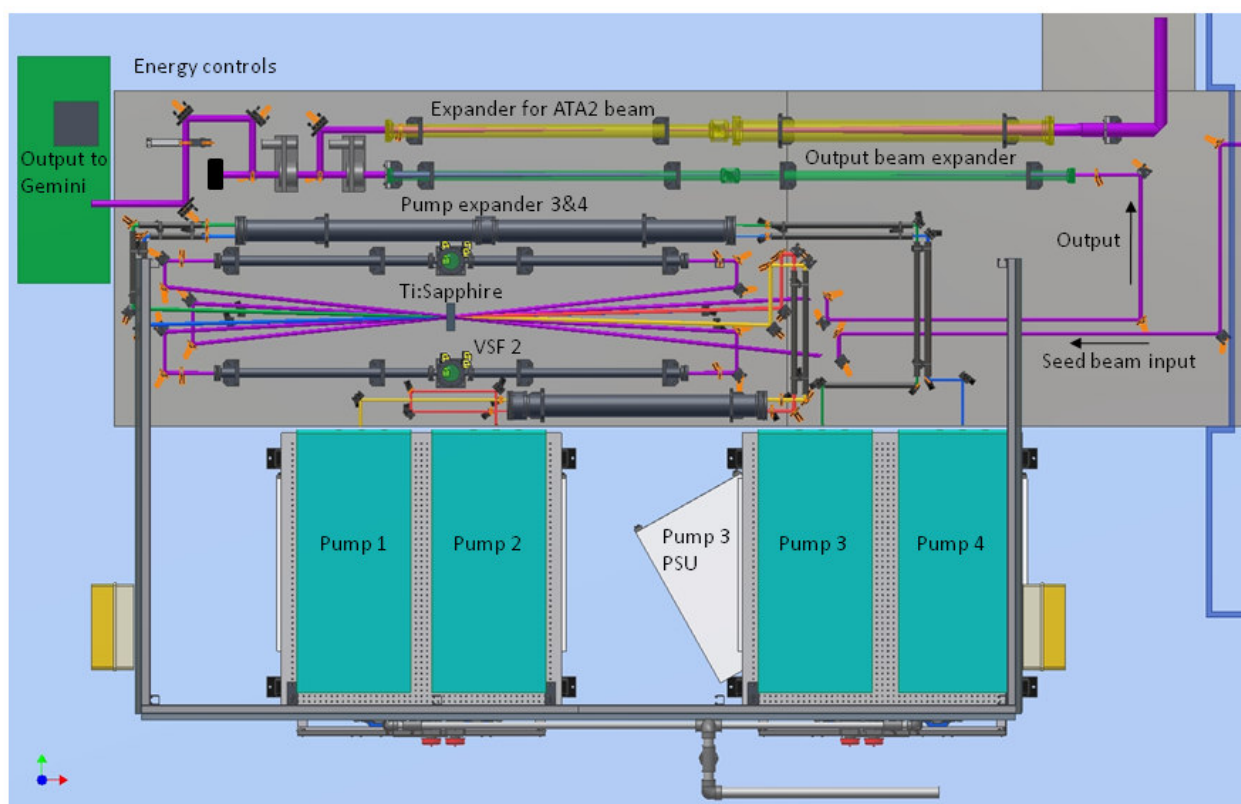


Figure 1. Schematic layout of new Astra Amplifier 3.

Triggering and timing

Each of the lasers requires a trigger to fire the lamps and another to fire the Q-switch. The requirement to adjust these independently and in groups led to a redesign of the scheme that was used for the Macholite. The new scheme uses three Stanford delay generators (DG535): one is the master delay box, and is triggered by a 10 Hz signal from the front end. Two of its outputs are used to trigger the other two boxes, which drive the four lamp triggers and the four Q-switches respectively, via their four main outputs. The Q-switch trigger channel is set up as the sum of two delays ($B + \text{constant}$); the B delay is zero for normal operation but is increased to 1 microsecond for alignment, so the crystal is thermally lensed

Environmental and beam enclosures

To protect the beams from air currents, some form of enclosure is essential. It is also necessary to protect the operators from the beams, especially the four pump beams which are a skin hazard as well as an eye hazard. The pump beams are enclosed in aluminium beam tubes as far as possible, as are the seed beam paths, but in the region around the crystal the overlap of paths makes this impossible. An environmental enclosure was designed, consisting of an extruded-aluminium framework with sliding and hinged Perspex panels, some with anodized aluminium skirts to block stray beams. The enclosure was installed only after the construction of the amplifier was

complete and the beams had all been aligned. A 50 mm wide gap was left along the top of the enclosure to allow cables and power leads to be fed through to equipment on the tables. Figure 2 shows part of the amplifier after the environmental screens had been installed: the four pump lasers are visible on the right, with the expander pipe for beams 1 & 2 and the smaller tube of the second VSF. The TiS crystal in its holder is visible on the left, along with two beam dumps on pneumatic slides which are used to dump the pump beams while the lasers are warming up.

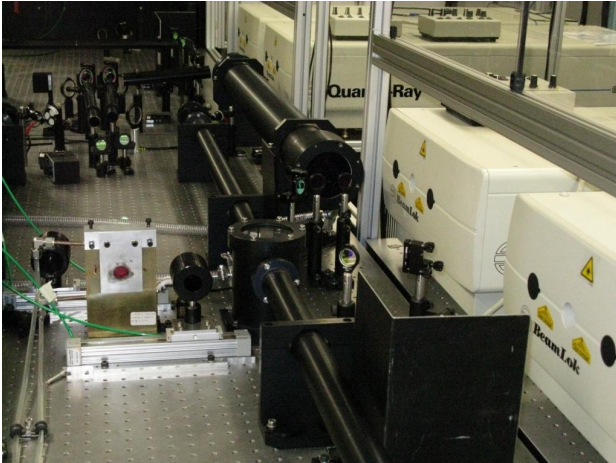


Figure 2. The new pump lasers and part of the completed amplifier.

Amplifier performance

The pump lasers were installed and commissioned in July 2009, and met their energy specification of 1.3 Joules per pulse at 532 nm. After a shakedown period, the alignment of the amplifier proved to be very stable from day to day, and the output energy was typically between 1.2 and 1.3 Joules with a reasonably uniform near-field profile (see Figure 3 below). The far-field shows a small amount of astigmatism; this is caused by the oblique passage of the beam through the thermally lensed crystal, and was also seen in the original design of the amplifier.

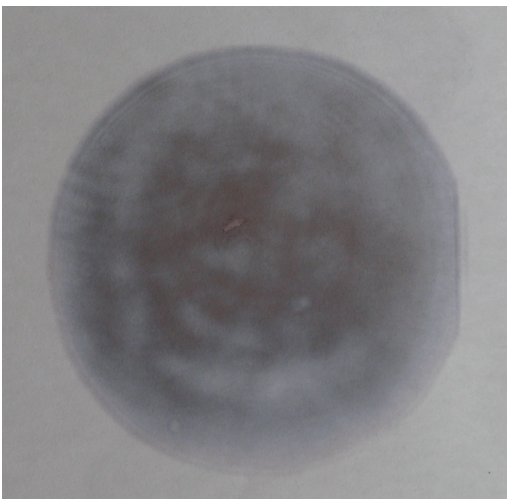


Figure 3. Burn pattern of 31 mm output beam.

New interlock system

While the upgrade of amplifier 3 and its infrastructure was in progress, the old PC-based interlock system was stripped out and replaced by a modern version using PLC technology. The old system was installed at the time the original Astra facility was set up, and some of the computers running the system

software dated from around 1999 or 2000. Concerns about reliability and the lack of spares made a compelling case for replacing them with a modern interlock system similar to the ones developed for Gemini, Astra Artemis and other areas of the CLF. As with all the modern systems, the user interface is via Magelis touch screens rather than mechanical switches and buttons, but authorization keys are still required and are issued to operators so they can gain entry to the areas and switch on hazardous devices. For the laser area entry door there are two parallel keyswitches, one of which is operated by the laser operator's key, and the other by a separate key for the users to enter the area when switching Astra off after operating during the evening. This second key cannot be used to switch on any of the lasers in the area, which provides an additional level of security against unauthorized operation of Astra. Linked to the interlock system is the display program, which runs the hazard display screens mounted outside the doors and at easily-visible places inside the areas. This new system covers the two Astra laser areas and also Target Area 2. There is no link to the Gemini interlocks, nor is one needed, because the interface between LA2 and LA3 consists of a single shutter in LA2. This shutter and its associated virtual shutter are controlled by the Gemini interlock system.

Conclusions

We have successfully completed a significant upgrade to Astra, which has removed one of the least reliable lasers from the system, and improved the layout and ease of access to the third amplifier. We have also replaced the increasingly unreliable PC-based interlock system with an up-to-date modern system that is compatible with similar systems elsewhere in the CLF.

Acknowledgements

The authors wish to thank the mechanical and electrical engineers of the CLF involved in this upgrade, whose efforts were vital in completing the work on schedule.

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Improved post-experiment data analysis at Astra Gemini

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Background

For the last few years Astra Gemini laser diagnostic data have been captured, processed and stored in a sizeable database. The “raw” data is already made available to users via NeXus¹ files and the eCAT and penguin applications (described in earlier years of the CLF annual reports^{2,3}), but with such a wealth of data available it is sometimes difficult to ascertain overall performance during the course of an experiment which can easily run to 1000 shots. We describe a recent addition to the penguin suite which aims to provide facility users (and laser operators) with a tailored digest of laser diagnostics to aid their own analysis.

Real-time data acquisition

The e-infrastructure set up for Astra Gemini ingests and stores a large amount of diagnostic data on a per-shot basis. The data currently comprises 500+ singular values (energies, pulse lengths, x- and y-positions, temperatures etc), 40+ images (compressed and uncompressed near- and far-field images, FROG images) and 200+ traces (spectrometer and scintillator traces), nearly 800 data streams in all. (There are also 20+ data streams of continuous data of vacuum pressures, crystal and ambient room temperatures, and pump energies.

Although specific requirements may vary for each experiment, there is usually a common subset of data channels of interest to the users. These can be chosen in consultation with the Principal Investigator and the data made available through the eCAT and/or penguin interfaces during experimental shots. Additionally the data, including singular values, trace images and camera images can be posted after every shot and made available as a web service. Figure 1 shows various laser parameters recorded for a series of shots in Comma-Separated Value (CSV) format. This CSV data can be downloaded and incorporated into analysis programs written for target area diagnostics. For example, a signal can be normalised to laser energy and plotted within seconds of the shot, enabling real-time assessment of data trends.

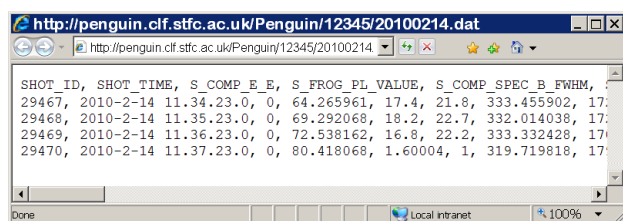


Figure 1: Extract from a CSV file.

Post-experiment analysis

The availability of laser performance data in a database allows CLF staff to carefully analyse the performance of the laser after an experimental campaign. This is crucial to pick up problems which may have been missed in day-to-day running. It also provides a valuable resource with which users can correlate experimental observations with deliberate or unintended changes in laser parameters. To this end, we are preparing reports in the penguin format which will be made available to the users soon after the end of the experiment. (These reports will not be posted to the public if there is any chance of a breach of confidentiality.)

Daily presentation of data

The data streams are presented as a shot-by-shot, day-by-day list of single values, traces and images. At the end of each day of experimental shots, singular value data streams are averaged and displayed following that day's shots. Traces are also averaged and displayed as a Scalable Vector Graphics⁴ (SVG) thumbnail which can be clicked on to enlarge and view in greater detail. The raw data for the plot is also available.

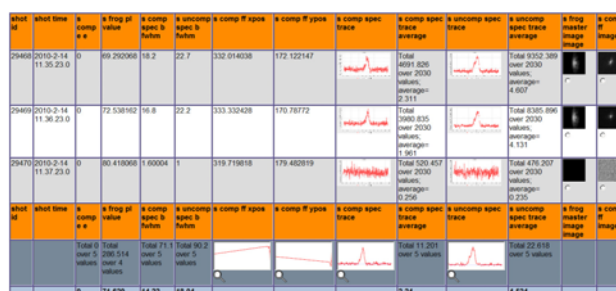


Figure 2: One day's shots extracted from the main display, showing raw and averaged data.

Summaries and representative data

The laser operator can subsequently view this data and, for image data streams such as far-field profiles and FROG traces, select the image which represents a typical image for that day. In this way we compile an overall summary table detailing, on a day-by-day basis, the average trace plots, the representative images and a scatter plot of X/Y position pairs. Following this is a table with the averaged singular data throughout the experiment and a list of all downloadable data (data, CSV and SVG files). The summary will be scrutinised by laser and target area operations managers, and any notable occurrences accompanied by a comment.

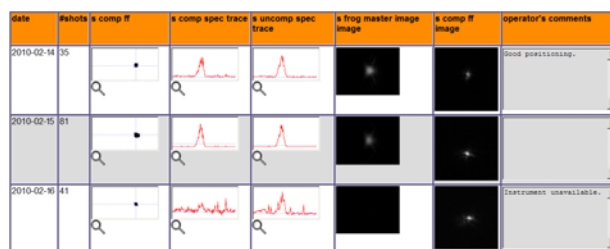


Figure 3: Three day’s worth of representative images and plots.

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A new phase (and amplitude) for Astra Gemini: Using a Spatial Light Modulator

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Introduction

Maintaining uniform spectral phase is essential in chirped-pulse laser systems to ensure that the pulse can be recompressed correctly at the end of the laser chain. In a system with a grating compressor, second- and third-order phase errors can (in theory) be compensated by adjustment of the grating separation and the angle of incidence. Phase errors that remain require other means of compensation, typically by an acousto-optic modulator such as a Dazzler. Spectral phase errors in the Astra/Gemini laser chain are at present corrected by means of a Dazzler positioned in the kHz preamplifier (Compact Pro). This device has several troublesome characteristics that reduce its effectiveness; in particular there appears to be a linkage between phase modulation and spectral modulation. If the amplitude of the phase correction exceeds a relatively small amount at any wavelength, or if the variation in applied phase correction with wavelength is too rapid, the result is a modulation of the spectral amplitude over the entire bandwidth. Many ultrashort-pulse laser systems use spatial light modulators (SLMs) to control spectral phase and amplitude independently. The advantage of such devices is that the light is dispersed at the point where the modulation is applied, which minimises the chance of a correction at one wavelength influencing what happens elsewhere in the spectrum. We report on the design and construction of an SLM-based phase correction arrangement for Astra Gemini.

Spatial light modulators

Spatial light modulators use arrays of liquid-crystal elements mounted between transparent electrodes. When a voltage is applied between the electrodes, the refractive index of the liquid crystal is altered, changing the optical path through that part of the modulator and thereby altering the relative phase of the light passing through it. If a broadband laser pulse is dispersed so that each element of the SLM array passes a small region of the spectrum, the relative phase of each part of the spectrum can be adjusted freely by applying different voltages to each element.

For our application we selected an SLM array made by Cambridge Research and Instrumentation, Inc (CRI), which has two liquid crystal cells mounted together between parallel polarizers. Each cell has 128 pixels of 100 microns width; the active part of the array is thus 12.8 mm across. The two cells are registered within 1 micron, so each pixel of the SLM array consists of two cells in series. The idea behind this is that it allows independent phase and amplitude control. The phase correction is determined by the average (or the sum) of the voltage values applied to the two cells of a particular pixel. If there is a difference between the voltages, this results in a rotation of the polarization and hence reduced transmission through the exit polarizer, thus modulating the spectral amplitude. The maximum phase difference that can be applied with this array is 3 radians, an amount that causes severe spectral modulation if applied with the Dazzler.

SLM optical configuration

For the SLM to work correctly, the light must be dispersed at the plane of the modulator, so corrections can be applied to narrow wavelength regions. The dispersion must then be

reversed to restore a collimated beam. The standard technique for doing this is a so-called '4F' system, in which the input beam, which is collimated, is dispersed by the first grating into a spread of directions. This fan of light is collected by an achromatic lens positioned one focal length F from the grating, so that the ray bundles of each wavelength are made parallel. Each bundle, however, is focused by the lens so that a well-resolved spectrum is formed at a distance F beyond it. This is the plane where the SLM is located, $2F$ from the first grating. Beyond the SLM, a mirror-image setup with another achromat and grating reverses the spatial chirp to restore an undispersed beam.

Our SLM has a width of 12.8 mm and a period of 100 microns; there are thus 128 active elements. Calculations based on a bandwidth of 100 nm full-width showed that a grating with 600 grooves per mm, combined with lenses of 200 mm focal length, would produce a spectrum that fitted comfortably within the SLM and used most of the pixels. Gratings of that groove density are readily available as replicas of ruled, blazed, gratings, with first-order efficiencies up to 90%, an important consideration to avoid loss of energy. Suitable achromats corrected for the near-infrared are available off-the-shelf from various suppliers, e.g. Thorlabs.

The SLM array is mounted in a large housing at a height incompatible with the 110 mm beam height of Astra. However, the modulator requires the beam to be horizontally polarized, and the gratings are also most efficient in the same polarization. Accordingly, the beam from the Astra pulse stretcher was raised to a height of 180 mm and turned through 90 degrees with a 2-mirror periscope, thereby changing the polarization and reaching a suitable height for the SLM. A second periscope after the 4F optics returns the beam to its original height and polarization and directs it into the original beam path. The setup was designed so that the SLM could be bypassed simply by moving the two lower periscope mirrors out of the path.

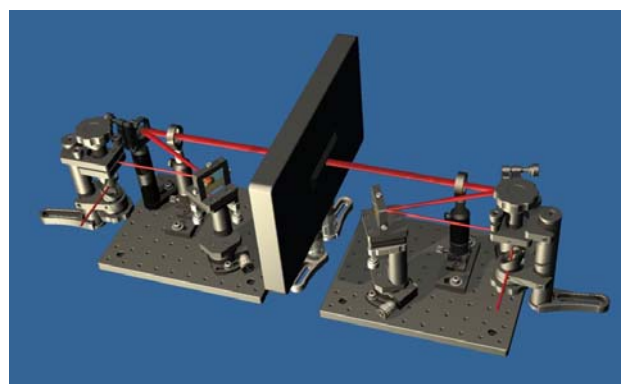


Figure 1. Design drawing of the 4F SLM optical setup

A schematic of the design is shown in Figure 1. The final setup is slightly different, in that the periscope optics are also mounted on the breadboards. The device is now bypassed by simply sliding the two breadboards towards one another by 20 mm: this allowed the 4F optics to be set up during the warm-up period of the laser each morning when the beam was not required, without disturbing the alignment of the laser in any

way. The cameras of the autoalignment system proved invaluable in ensuring that the 4F setup was adjusted correctly, because the image sizes and positions are very sensitive to errors in beam collimation and pointing. One issue that was noted is that the 4F optics invert the beam: this has the effect that the autoalignment system becomes unstable, as a correction applied before the 4F setup causes the spot to move away from the reference position on any camera downstream of the SLM. When the SLM is finally made a permanent part of the system, the auto-alignment software will have to be 're-trained'.

Control program and user interface

The SLM is controlled via ASCII commands using a high-speed USB interface. Two 128-pixel masks are available, which can be controlled independently to effect phase and/or intensity modulation. Figure 2 below shows the screen of the user interface.

Firstly, the operator needs to define the masks.

Polynomial phase settings: Four polynomial phase settings are available: K_1x , K_2x^2 , K_3x^3 and K_4x^4 . These can be controlled independently using sliders and are plotted separately in the top graph. Their combined values are shown in the combined phase pattern plot below it. All values are taken modulo 2048; i.e. they are wrapped if their values go outside the range ± 2048 .

Local phase settings: These are defined by small groups of pixels (usually in groups of three) and defined using the two bitwise sliders. To create a phase hole, the operator chooses the position with the horizontal slider and the amplitude with the vertical one. A Gaussian function is used to smooth the curve. These local corrections are summed, then plotted both separately and combined with the polynomial values. For pure phase corrections the same voltage is applied to each mask.

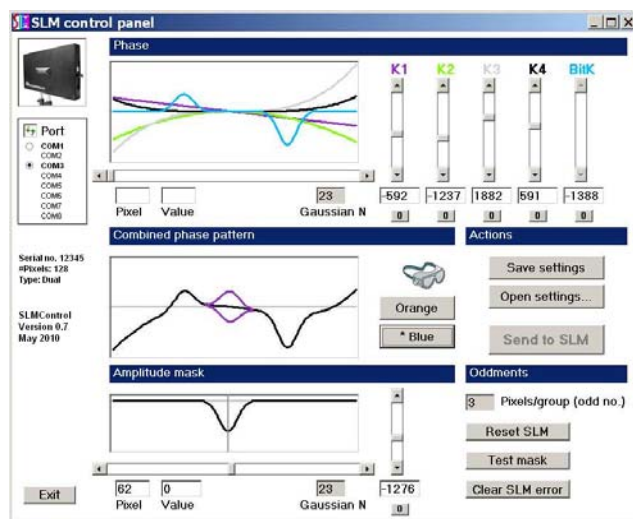


Figure 2. Screen image of the SLM user control interface

Amplitude settings: These are defined at the bitwise-level using the lowest plot and two sliders, and by applying a Gaussian function. The voltages applied to each mask are adjusted to the required difference, so that their combined value (which controls the phase correction) is unchanged. The effect is shown on the combined plot. In this way a spectral hole of any desired width and depth can be created.

Once the operator has defined the masks to be used, they can be downloaded to the SLM itself using the "Send to SLM" button. Settings can also be saved to and restored from XML files.

One complication was the requirement to be able to use the SLM interface while using safety goggles which restrict visibility as well as colour range. There are two types of goggles in common use, one transmitting in the orange part of the spectrum and one in the green-blue. This was solved using

two sets of colour tables and a button to swap between them. Both versions appear slightly unusual to the naked eye, but more importantly they are usable wearing goggles in a laser laboratory with high-power laser hazards present.

Results and further work

Only brief preliminary tests have been carried out so far. However, the device was shown to be capable of modifying the spectrum from Astra with a resolution significantly better than the Dazzler. The depth of modulation was essentially 100%, i.e. a chosen part of the spectrum could be completely blocked. We have not yet had an opportunity to try controlling the phase at the output of Gemini using the device. The intention is to retain the Dazzler, primarily as a means of controlling the spectral bandwidth from the front end. Assuming the SLM works as expected, it will become the primary means of phase control, and a secondary means for adjusting the shape of the spectrum that enters the Astra amplifier chain. There is also an option in the SLM to store more than one set of masks, and to switch between them if the laser is running in alternating single- and double-stretch mode.

Ultrafast gated imaging of laser produced plasmas using the optical Kerr effect

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Introduction

Optical probing is a powerful diagnostic tool which is frequently used for high energy density plasma experiments with high power lasers. It enables the measurement of plasma electron density^[1] and visualization of energy transport and shock propagation [e.g. Ref. 2]. Short (<1ps) laser pulses can be employed as a backlighter for transverse imaging of the interaction with a temporal resolution limited by the transit time of the pulse through the plasma. The emission of light created by the laser-matter interaction can also be imaged: both the nanosecond timescale radiation from the directly heated plasma and also short bursts of harmonic or transition radiation emitted as relativistic electrons leave the target^[3]. The resolution for these measurements is determined by the exposure time of the camera, which can be electronically gated to be on the timescale of ~100 picoseconds. The gate can be timed to eliminate the majority of the self-emission from the plasma which causes a problematic background in high intensity experiments. The detector response can be made faster by driving the gate optically rather than electronically by incorporating into the imaging system an optical Kerr gate (OKG). This is composed of two crossed polarizers around a non-linear medium^[4] which is opened by passing an intense laser pulse through the gating medium to introduce a polarization rotation to the probe pulse allowing it to reach the detector. The OKG is used extensively for spectroscopic applications^[5-8] and is a standard technique within the LSF. It has also been employed for ballistic-photon imaging^[9-11], laser contrast improvement^[12] and is the basis for polarization gated frequency-resolved optical gating used to diagnose laser pulses^[13]. Carbon disulfide is commonly used for the gating medium because of its strong non-linear response but it has a slow relaxation time ~1ps. For ultrafast applications, glass or crystals must be used because of their much faster response^[6,7] but these usually need higher laser intensities to induce the necessary birefringence.

Design of the Kerr gated imaging system

We report on the application of Kerr gating to the imaging of laser-produced plasmas on Astra^[14,15]. We present shadowgraphic images of the laser interaction with a solid target to demonstrate the suppression of background light from plasma self-emission, which otherwise dominates the image. We also discuss the exceptional performance that should be achievable with further optimization of the imaging system. The gated imaging scheme comprised a four lens system which image relayed the plasma interaction to the position of the OKG and then re-imaged with magnification to the charge-coupled device (CCD) camera. A linearly polarized probe beam was used as a backlighter, which was split before the CCD and focused onto a photodiode to monitor gating performance. Short pass filters were used to block light at the fundamental 800nm wavelength. The OKG was formed by aligning a collinear gating beam with its polarization oriented at 45° to the probe through a non-linear medium and placing a polarizer in front of the detectors. The experiment was performed in Astra Target Area 2 using the output from the hollow fiber pulse compressor (HFPC)^[16,17] to provide a good spatial profile. The fiber was operated without a gas fill, preventing spectral broadening and

so the pulse duration remained at ~60fs. A second harmonic (400nm) probe pulse was generated in a thin β -Barium-Borate crystal and the remaining 800nm light was used as the gating beam. This was focused to 1mm diameter at the OKG, generating sufficient intensity to drive the gate while remaining large enough to provide an adequate field of view for plasma imaging.

Characterisation of the Kerr gate

We investigated the properties of the gated imaging system by measuring the transmitted 400nm signal. To ensure the entire probe beam experienced polarization rotation, its size at the OKG was made smaller than that of the gating beam. We tested two glasses as gating materials: fused silica (FS) and Schott NSF-66. These samples were 1mm thick, 1 inch diameter windows and the FS had a broadband antireflection coating. Both glasses are inexpensive and FS is well known to have an instantaneous (~1fs) response^[13]. The high-index NSF-66 (1.914 at 632.8nm) was chosen because the effect is expected to be stronger with increasing linear refractive index^[6].

The signal as a function of the gating beam intensity is shown in the inset to Fig. 1 for FS. The Kerr effect becomes significant once the intensity exceeds $8 \times 10^{10} \text{ Wcm}^{-2}$ (150 μJ) and follows a quadratic dependence with increasing energy (as seen in Ref. 4) until $\sim 2.5 \times 10^{11} \text{ Wcm}^{-2}$ (500 μJ). The transmission is ~20%, which seems reasonable since the intensity required to drive a half-wave polarization rotation of the probe is $\sim 7.5 \times 10^{11} \text{ Wcm}^{-2}$. In NSF-66, the maximum intensity was limited by the onset of white light generation by the gate pulse causing saturation of the CCD camera. At this intensity, the efficiency of NSF-66 was only slightly higher (~23%) than the FS.

The transmission for both materials is plotted against the gating beam delay in Fig. 1. In NSF-66 the birefringence endures for ~1.5ps precluding ultrafast gating. In contrast the FS gate

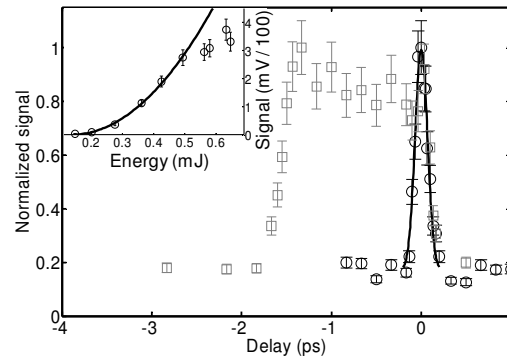


Figure 1. Probe signal as a function of gating beam delay using as the Kerr medium fused silica (black) and NSF-66 (gray). The inset shows probe signal versus gating beam energy for the fused silica gate.

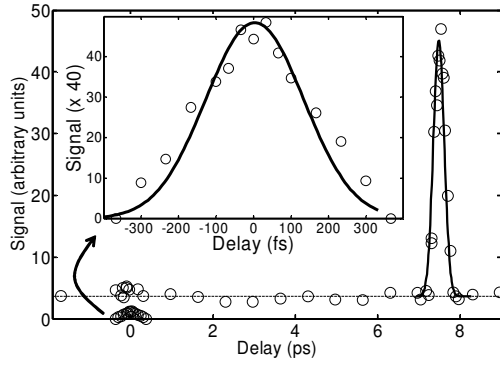


Figure 2. Gated measurement of a strong (7.5ps) and a weak (0ps) probe pulse. The dashed line indicates the background level caused by the strong pulse. The inset shows the weaker pulse measured with no strong pulse present (signal x 40).

transmission is fitted with a full width half maximum of 180 fs. Given that FS is known to have $\sim$ fs response time the duration of the gate should be limited only by the gate duration of $\sim$ 60fs. However, since the gate is short, the signal width that we measure is determined by the duration of the probe pulse which is lengthened because of transit through $\sim$ 8mm of glass. In future work we intend to conduct a more sophisticated measurement of the gate duration.

We measured the dynamic range of the system by splitting the probe and attenuating one pulse by a factor of 50. Figure 2 shows a scan of the gating beam delay with the zero position defined as the peak of the weaker pulse. The attenuated pulse could be resolved in the absence of the stronger pulse (at 7.5ps), when the background level was negligible. With the stronger pulse present the background level increased to $\sim$ 40mV (dashed line), almost obscuring the weak pulse. This $\sim$ 7% background is caused by leakage of the strong pulse through the crossed polarizers which, in this arrangement, had an extinction ratio of 100.

Application of Kerr gating to plasma imaging

Adding the OKG to the imaging system suppresses the level of plasma emission since this occurs on a nanosecond timescale compared to the $\sim$ 100fs timescale of both the gate and the probe beams. In high intensity interaction experiments this background is problematic because it necessitates a correspondingly bright probe beam. The use of gated imaging relieves this requirement. We demonstrated background reduction by recording shadowgraphic images of plasma formation in a 100 μ m thick glass target irradiated at $\sim 2.5 \times 10^{14}$ Wcm $^{-2}$ by 2mJ, 60fs laser pulses focused to a 70 μ m spot. At this intensity the single-shot emission was not bright compared to the probe beam so we used multiple laser shots to create damage craters with complicated structure which lead to greater laser absorption^[18]. A series of 100 laser shots was incident onto a thin glass target (a movie of this is available on request). Without gating after about 20 shots the self-emission became much brighter and saturated the camera, obscuring the interaction region. In contrast, the use of the OKG enabled clear imaging of the structure of the glass and diagnosis of the emergence and propagation of cracks. Recording a sequence of 250 shots allowed us to observe the laser drilling completely through the target (movie available). The results are shown in Fig. 3 which shows irradiation of the crater after 25 shots (a) without and (b) with optical gating, with a probe delay of 2.5ns. In (a) the interaction region is obscured by self-emission whereas this background is not seen on the gated detector in (b).

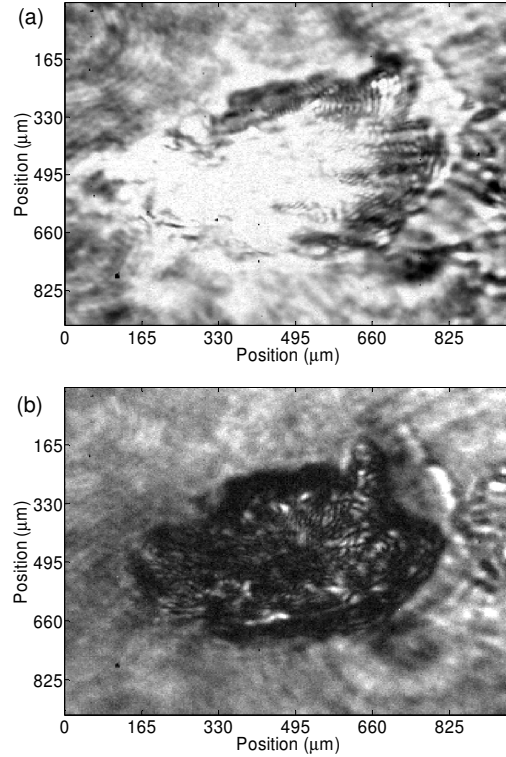


Figure 3. Shadowgraphic images of a glass target irradiated at laser intensity 2.5×10^{14} Wcm $^{-2}$ at a probe delay of 2.5ns (a) without and (b) with optical gating. The target has previously been subjected to 25 laser shots to enhance plasma self-emission.

Alternative gating materials

As well as the two glasses we tested two alternative gating materials which have previously been used in fast-response OKG systems: zinc oxide^[19] and lead-bismuth-glass (LBG)^[12,20]. When we used ZnO we found that rather than inducing the polarization rotation necessary for gating, the intense gating pulse induced a strong absorption of the 400nm probe pulse as shown in Fig. 4. This makes it inappropriate for Ti:sapphire second harmonic probing but it should be usable for other probe wavelengths. Importantly, the use of this ZnO wafer had no detrimental effects on the image quality, Fig. 5(a), so we can employ different crystalline materials^[7] for 400nm

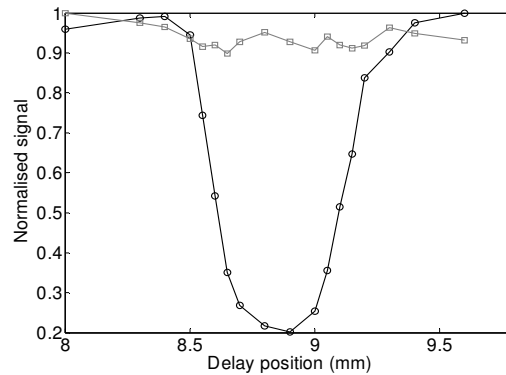


Figure 4. Probe signal (black) as a function of gating beam delay using ZnO as the Kerr medium with fluctuations in the infrared pulse energy shown (gray).

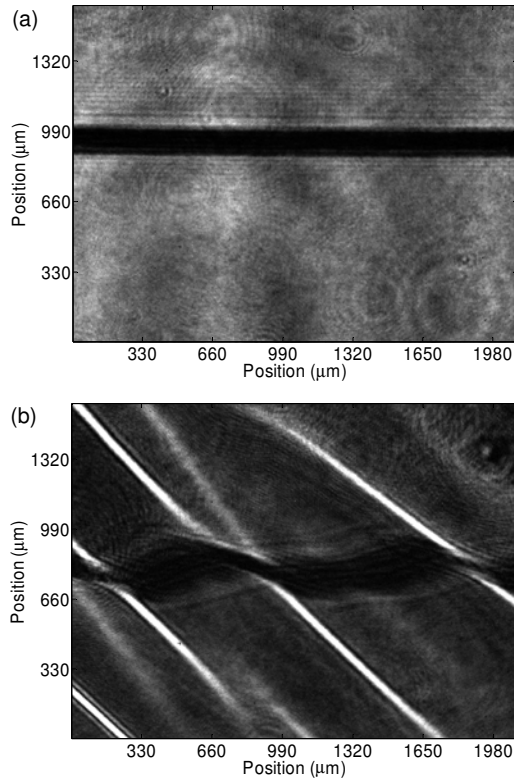


Figure 5. Imaging of a wire through (a) zinc oxide wafer and (b) lead-bismuth-gallium glass. Structure in the glass prevents the use of this sample for an imaging application.

probing. This should be compared to the image in Fig. 5(b) obtained using the available sample of LBG, which introduced striations to the image highlighting the need for good quality glasses for these imaging applications.

Conclusions

We have demonstrated that high speed optical Kerr gating can be incorporated into the imaging systems commonly used for diagnosing high energy density plasmas created using intense laser pulses. We have presented images which show the effective elimination of plasma self-emission from the camera. This is of great benefit for experiments using ultra-intense laser systems in which this background constitutes a common problem. These images also highlight the potential of the optical gating technique for investigations of material properties, such as in situ laser optic damage testing^[21]. The gating beam can be tilted with respect to the probe^[13,22,23] to convert the device to an ultrafast all-optical streak camera. These instruments will thus constitute low-cost plasma diagnostics with substantially higher dynamic range and superior temporal resolution than current generation electro-optic devices.

In principle, the gated system should be capable of an instrument response on a sub-10fs timescale and optimizing the performance remains a challenge requiring further research. It should be noted that even without an ultrafast response, optical gating on the picosecond timescale, using conventional gating materials (CS_2) and low intensities (5 GW cm^{-2}) can be used for background suppression in high intensity experiments. We aim to introduce optically gated imaging as a standard diagnostic for high power laser experiments within the CLF.

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Astra laser operators and target fabrication team. Sections of this report and figures 1 – 3 & 5 were adapted with permission from Ref. 14, ©2010, American Institute of Physics doi:10.1063/1.3279139.

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Two-photon fluorescence correlation spectroscopy: FCS

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Introduction

Fluorescence correlation spectroscopy (FCS) is a powerful single-molecule detection technique used by physicists, chemists, and biologists to characterise the dynamics and interactions of fluorescent species by recording and correlating their fluctuations in fluorescence intensity within a microscopic detection volume^{1,2,3}. The spontaneous intensity fluctuations arise from the diffusion (Brownian motion, active transport...) of a few molecules in and out of a restricted sub-micron observation volume defined by a focused laser beam in the sample ($\sim 1 \text{ fL} = 10^{-15} \text{ L}$), as well as from any other processes (chemical reactions, photodynamic process, and conformation change) which convert the optical species between states with different emission properties. FCS can provide dynamic information of simultaneous processes occurring on different time scales (from nanosecond to microsecond). In a typical FCS measurement, fluorescence intensity is recorded for a small number of molecules in a specified volume over a time range (generally between 20-60s), and the fluorescence intensity is then analysed in terms of its temporal autocorrelation function $G(\tau)$ defined by

$$G(\tau) = \frac{\langle \delta F(t) \cdot \delta F(t + \tau) \rangle}{\langle F \rangle^2} \quad (\text{figure 1}).$$

The autocorrelation

function contains information about equilibrium concentrations, reaction kinetics and diffusion rates of molecules in the sample (2D, 3D, anisotropic diffusion, etc.).

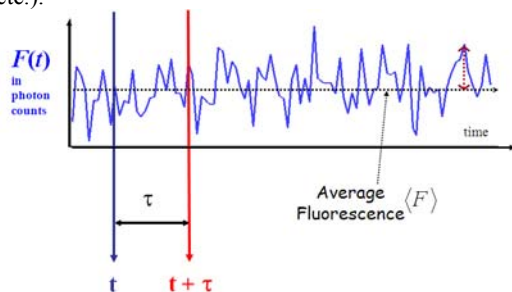


Figure 1: Calculating the autocorrelation function.

Two-photon excitation (TPE), where the fluorophore is excited by simultaneous absorption of two photons, has been applied to FCS for reasons similar to those that have motivated its use in fluorescence microscopy-inherent spatial confinement of excitation, diminished photobleaching and phototoxicity, less scattering, and better optical penetration in turbid media. Moreover it offers the possibility to simultaneously excite several

spectrally distinct dyes with a single wavelength for dual-color cross-correlation measurements.

Results

To demonstrate the potential of this technique using our TiSa confocal setup, the diffusion coefficient of the Rhodamine dye was measured for different concentrations and laser powers. Assuming a Gaussian shape for the confocal volume³, the simplest autocorrelation function for a 3D diffusion is given by:

$$G(\tau)_{3D} = 1 + \frac{G_{\text{triplet}}(\tau)}{n} \frac{1}{\left(1 + \frac{\tau}{\tau_{3D}}\right) \sqrt{\left(1 + \frac{\tau}{\tau_{3D} S^2}\right)}} \quad \text{with}$$

$$G(\tau)_{\text{Triplet}} = \left(\frac{1 - T + T \cdot \exp(-\tau/\tau_{\text{Tri}})}{1 - T} \right)$$

where T denotes the fraction of molecules in the triplet state, τ_{Tri} the triplet lifetime, and n is the number of molecules inside the probed volume. S is the shape parameter of the confocal volume, the ratio between the axial to the radial volume: $S = \frac{\omega_z}{\omega_{xy}}$.

τ_D the diffusion time gives the diffusion coefficient D :

$$D = \frac{\omega_{xy}^2}{8\tau_D} \quad \text{for a two-photon absorption process.}$$

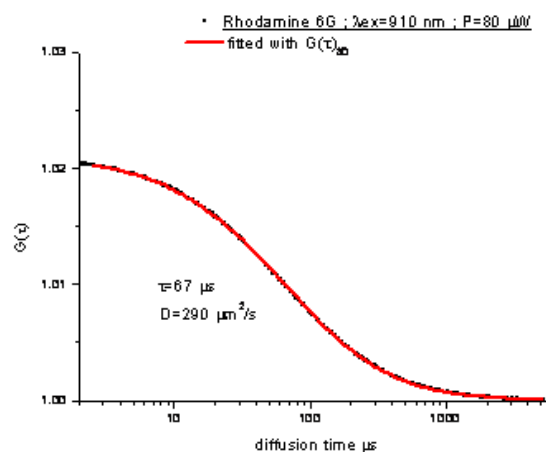


Figure 2: Measuring the diffusion coefficient of Rhodamine 6G in aqueous solution.

The fit (figure 2) gives access to the coefficient of diffusion of the dye ($290 \mu\text{m}^2/\text{s}$), the concentration of the solution (2.6 nM) and the triplet lifetime (900 ns). The initial amplitude of the autocorrelation function $G(\tau=0)$ varies with the inverse of the number of molecules inside the detection volume: the concentration of the solution can be probed (figure 3).

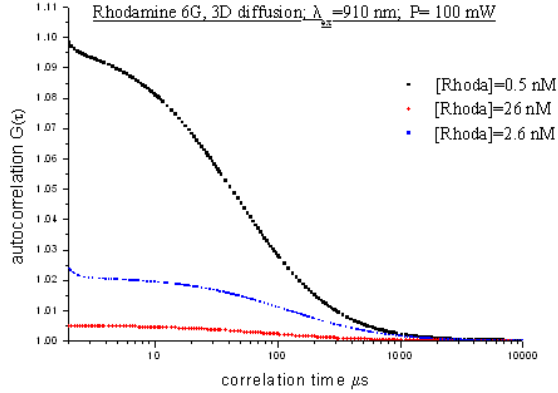


Figure 3: The effect of the particles concentration on the autocorrelation function.

If the laser is too powerful, erroneous values for the initial concentration (photobleaching of the dye) and for the diffusion time may result (figure 4).

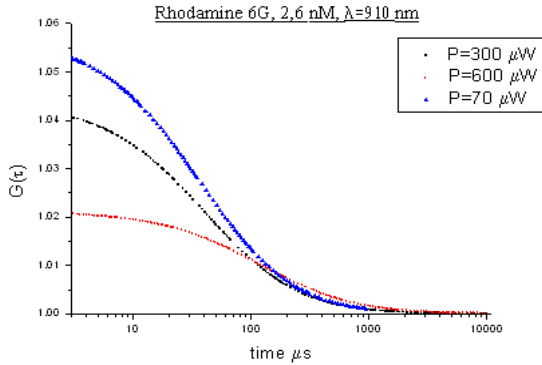


Figure 4: The effect of the laser power on the autocorrelation function.

In figure 5, FCS has been used to measure the mobility of poxvirus inside a live cell using a 2D diffusion model for the data fitting (surface diffusion):

$$G(\tau)_{2D} = 1 + \frac{G_{\text{triplet}}(\tau)}{n} \frac{1}{\left(1 + \frac{\tau}{\tau_{2D}}\right)}$$

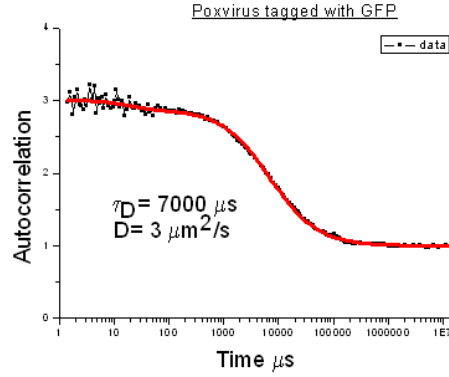


Figure 5: Diffusion of poxvirus inside cell.

With our setup, we can use cross correlation to remove detector noise (after pulse signal), or to probe interactions between two different fluorescent labels with distinct excitation and emission properties.

Conclusion

Fluorescence correlation spectroscopy (FCS) is a technique used for spatial and temporal analysis of molecular interactions in solution (in vitro and in vivo). The power of FCS lies in its capability to extract dynamics (molecular diffusion, dimerization, complex formation, triplet lifetime...) over a wide range of time scales from sample concentrations ranging from subnanomolar to micromolar. Renewed interest in FCS in recent years has been stimulated by the fact that it is inherently miniaturised and therefore applicable for high-throughput screening applications⁴.

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The ULTRA time-resolved IR, 2D-IR and T-2D-IR station

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Introduction

Ultrafast time-resolved pump-probe spectroscopies are now widely applied techniques for the detection of short-lived reactive intermediates and excited states and provide valuable information on structure and dynamics of molecular species in the condensed phase.^{1,2,3}

Over the last three years the LSF has developed the ULTRA facility, the successor to the PIRATE⁴/Kerr-gated Raman⁵ system, which along with transient absorption and femtosecond stimulated Raman spectroscopies is optimized for time-resolved infrared investigations of biological species. The system is highly sensitive being capable of measuring changes in sample absorbance of ΔOD ca 10^{-5} in 1 second. Along with the high sensitivity the instrument is also extremely flexible for multiple beam experiments such as ground state and transient two-dimensional infrared spectroscopic measurements.

Instrumentation

The system is a custom designed 10 kHz dual titanium sapphire amplifier (Thales Laser) with single oscillator seed source (FemtoLaser). One amplifier produces 40-80 fs pulses while the other produces 1-3 ps pulses both at 10 kHz, 0.8 mJ and 800 nm. The two are synchronized to < 70 fs through the sharing of a single seed source; long term drifts in the timing of the two amplifiers, on the picoseconds timescale, are controlled using feedback to the picosecond regenerative amplifier cavity which contains a piezo stepper motor. This feedback control is vital for transient-2DIR data acquisition where the timing of the fs UV pump and IR probe and ps IR pump must all be kept to sub-ps accuracy.

At their outputs each amplifier is split into two to pump computer-controlled OPAs (TOPAS, Light Conversion) equipped with harmonic units, and sum frequency and difference frequency units. This combination allows wavelengths in the range 200 – 15000 nm to be accessed with either fs or ps pulse lengths. The polarization of all the output beams can be set using either computer-controlled zero order or broadband wave plates. The combination of fs and ps pulse lengths and wide tuning range provides enormous scope for a range of pump-probe techniques including time-resolved infrared (TRIR) spectroscopy and ground state and time-resolved double-resonance two-dimensional IR spectroscopy (2D-IR and T-2D-IR). All these studies employ the fs IR beam, which has a bandwidth of ca 500 cm^{-1} , as the probe and use either the fs UV-vis, and/or the ps IR beam with its narrow, 12 cm^{-1} , bandwidth as the pump.

A pump-probe time-delay is introduced and controlled using an optical delay line (Newport), and the pump beam repetition rate reduced using optical choppers (TTi). The repetition rate of the pump is technique dependent, for TRIR and 2D-IR 5 kHz is used and for T-2D-IR 5 kHz and 2.5 kHz are used for the UV and IR pumps respectively. The pump and probe beams are

then focused into a temperature-controlled (-60 to $> 80\text{ }^{\circ}\text{C}$) sample cell (Harrick Scientific, $6\text{ }\mu\text{m}$ to 1 mm pathlength) with typical beam diameters of ca 100 and $50\text{ }\mu\text{m}$ respectively. To reduce photo-degradation samples are either rastered in the x,y plane or rastered and flowed with a minimum flow volume of ca $300\text{ }\mu\text{l}$. The probe beam is then split into two with each half focused into separate spectrographs (0.25 m f/4 DK240, Spectral Products). Light is then detected using 128 element mercury cadmium telluride (MCT) arrays (IR Associates) which are read out by a custom DAQ system (QD).

The spectral window is controlled along with the resolution by using different diffraction gratings and use of the detectors which can either be used as 128 element probe and 128 element reference or 240-250 element (allowing for some spectral overlap) probe by combining the two arrays with the spectrographs tuned to different wavelength regions. The dual-detector approach produces an unsurpassed spectroscopic window of ca 500 cm^{-1} . The major influence in background variation in the difference spectra is due to spectral fluctuations which lead to either baseline slope or offset changes. When dual probe detectors are used a 64 element MCT array (Infrared Associates) is used as the reference and is spectrally matched using a combination of interpolation and lower density grating. This referencing allows spectral changes of the order of ΔOD ca 10^{-5} in 1 second to be recorded.

The first element of one of the MCT probe detectors is reserved for monitoring IR pump intensities, throughout a 2D-IR spectrum scan, providing the relative pulse intensity data.

The whole of the IR spectroscopy system is purged by nitrogen gas to remove interference from atmospheric water vapour and carbon dioxide which have large absorptions in the ca 1600 cm^{-1} and 2400 cm^{-1} regions.

The data acquisition and system is controlled through a NI LabView program written in-house. This also controls the scanning of the IR pump beam for the 2D-IR and T-2D-IR experiments and the tuning of the probe OPA. Further details on this can be found in another article in this annual report.

Further information on the instrumentation can be found in reference ⁶.

Results

To demonstrate the broad bandwidth of the instrument the TRIR spectrum of 4-phenylbenzophenone in CD_3OD was recorded, Figure 1, as a comparison to previously recorded data.⁷ The spectrum, which was recorded with a 5 pixel overlap between the two probe detectors, clearly shows the $> 500\text{ cm}^{-1}$ bandwidth - ca 4 times that available on the previous PIRATE system.⁴

The negative peaks in the TRIR spectrum of 4-phenylbenzophenone correspond to the loss of the ground

state parent species upon excitation and the positive peaks to the new species formed. In this case two species are produced the singlet excited state, which has peaks at ca 1460 and 1554 cm^{-1} , and the $\pi\pi^*$ triplet excited state which is responsible for the remaining 7 features. Figure 2 shows the kinetics of the decay of the singlet state leading to the formation of the triplet state which has a rise time of ca 15 ps.

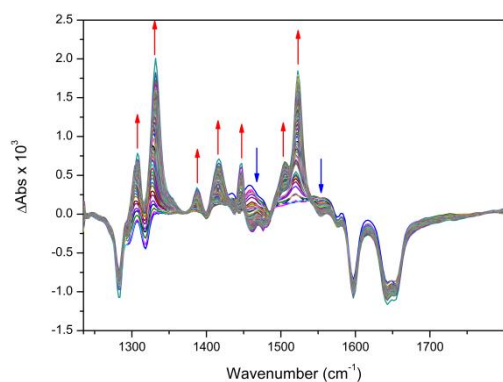


Figure 1 TRIR spectra of 4-phenylbenzophenone in CD_3OD obtained at 1 to 800 ps following irradiation (267 nm, 3 μJ , magic angle polarization) and 100 s data acquisition. Blue arrows mark the decay of the singlet excited state while the red arrows mark the formation of the triplet state.

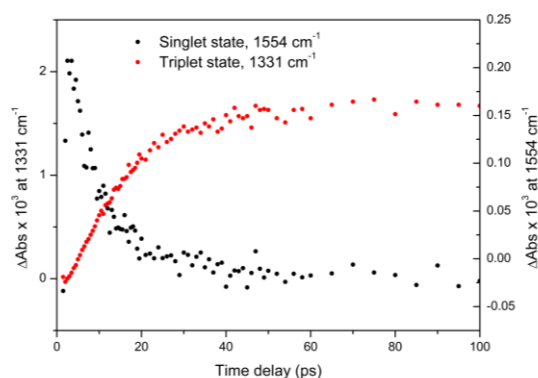


Figure 2 Kinetics of the decay of the singlet excited state of 4-phenylbenzophenone in CH_3OD at 1554 cm^{-1} (filled circles) and rise of the triplet state at 1331 cm^{-1} (open circles).

Figure 3 illustrates the 2D-IR technique, using 5'-GMP in D_2O buffer as an example; for reviews of the technique see references 8 and 9. Here the 1D-IR spectrum lies along the pump-probe frequency diagonal, peaks **1** and **2** are assigned to the carbonyl stretch at 1662 cm^{-1} and ring mode at 1585 cm^{-1} respectively. The main features of interest in 2D-IR spectroscopy are the features that lie off this diagonal, **3**, where pumping of one band at *eg* 1662 cm^{-1} has a significant effect on other vibrations through vibrational coupling. The bleaches (in blue) correspond to loss of the $\nu = 0-1$ mode on excitation and the transient absorptions (in red) are those of the $\nu = 1-2$ species formed. Spectra recorded at each individual pump wavelength show a slice through the 2D spectrum and the 2D plot is composed of all slices.

An example of the T-2D-IR spectroscopy can be found elsewhere in this report in the article by Dr Neil Hunt.

Conclusions

We have demonstrated the potential of the ULTRA infrared station which can be applied to a wide range of techniques – TRIR, 2D-IR and T-2D-IR, examples of which can be found elsewhere within this report. The instrument is a significant improvement over the PIRATE facility having ca 4 times the bandwidth (ca 500 cm^{-1}) enabling spectra to be recorded over a

wide spectral window in shorter time and with reduced sample quantities, a feature that is particularly important when studying valuable biomolecules. The instrument is aided by advanced control software enabling scanning of the 2D-IR pump wavelengths, delay lines and waveplates permitting many experimental variations to be recorded in a single data file.

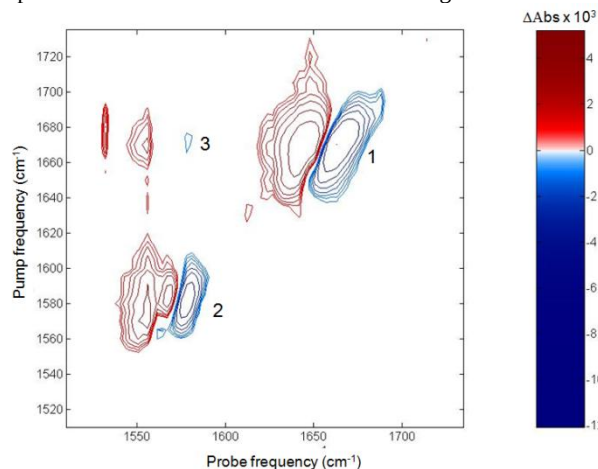


Figure 3 2D-IR spectrum of 5'-GMP (10 mM) in D_2O buffered solution (50 mM phosphate buffer) recorded 2 ps after excitation (ca 1 μJ , magic angle polarization).

Acknowledgements

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Developments within the EPSRC Laser Loan Pool

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Introduction

Throughout 2009/10 the Laser Loan Pool continued to provide laser systems to the UK research community providing a cost effective, valuable resource aiding many research groups and helping in the training of a number of postgraduate students.

Research projects undertaken with the help of the Loan Pool range from magneto-optical data storage to polymer dynamics.

Developments

During the year a number of changes have occurred, the most significant of which is the establishing of a Steering Committee for the facility. This committee consists of four UK researchers with differing areas of expertise, spanning the fields of chemistry, physics, bioscience and materials/engineering research. The committee members are:

- Prof Lin Li – University of Manchester
- Prof Pavlos Lagoudakis
- Dr Andrew Hudson – University of Leicester
- Dr Michael Hippler – University of Sheffield



Members of CLF staff and the Loan Pool steering committee at the January meeting. From left to right, Tony Parker, Pavlos Lagoudakis, Lin Li, Andrew Hudson, Michael Hippler, Mike Towrie and Ian Clark

The first committee meeting was held in January 2010 and was highly productive with a number of key points raised. Research fields that could be better supported by the facility were identified, namely materials and bioscience; in addition the possibility of supplying whole experimental set-ups, for *eg* spectroscopic techniques, was considered but at present is unlikely.

The potential benefits to the community of a wide range of new laser systems were discussed and a number of possible purchases identified. These include a supercontinuum source with acousto-optic tunable filter (AOTF), a femtosecond tunable titanium:sapphire (Ti:S) oscillator with optical parametric oscillator (OPO) and a picosecond high power laser for cold ablation type machining.

Tunable Ti:S oscillator and OPO

A tender exercise is currently in progress for the purchase of this system. The emphasis for this system is ease of use *ie* turn-key, hands-free and computer controlled. This system is expected to appeal to researchers in the fields of physics and bioscience and is designed with coupling into a microscope in

mind. Applications are likely to include coherent anti-Stokes Raman spectroscopy (due to the tunable oscillator and OPO), multi-photon excitation microscopy and materials processing. This system is hoped to be in place by December 2010.

Supercontinuum source with AOTF

This laser system will increase the scope of the Loan Pool further having a wide range of potential applications to support research in the fields of physics, chemistry and the life-science interface *eg* single molecule spectroscopy and optical coherence tomography.

This laser has been purchased and is a quasi CW single-mode supercontinuum white light laser from NKT coupled with acousto-optic tunable filter; specifications are provided in the table below. Delivery is expected in early August 2010.

Source	SuperK G2 Extreme
Tunability	460-2400 nm
Total average power	> 2 W
Spectral density	2.4 mW/nm max, 0.8 mW/nm min 465-750 nm 2.0 mW/nm max, 0.3 mW/nm min 750-1100 nm 1.2 mW/nm max, 0.3 mW/nm min 1100-2000 nm
Repetition rate	40 MHz
Master source pulse length	5 ps
Polarization	Unpolarized
Ancillaries	SpectraK dual AOTF with single visible AOTF (450-750 nm) and second channel for direct infrared output. Armoured fiber & collimator

In addition to the above specifications the system has a modification in the form of a second amplifier module to permit access to *ca* 100 mW of spectrally clean 1064 nm.

High power laser

This laser would be for the materials and engineering community. However when the necessary ancillary items are taken into account this would be a very costly system and as yet a definite decision on the purchase remains to be made. The Loan Pool would very much welcome the thoughts of the community as to whether this system is desired.

Conclusions

Two new laser systems are to be introduced to the facility by year end with a third under consideration. As such some lasers will be retiring, these will be the current CWL1 argon-ion laser and one of the Nd:YAG pumped dye systems (NSL5), a call for applications for these systems will go out in late summer 2010.

Acknowledgements

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The OCTOPUS Imaging Cluster: A New Imaging Facility in the Research Complex at Harwell

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Introduction

The past year has again seen major changes in the Lasers for Science Facility (LSF). The LSF's operations in the biological imaging area were relocated to the Research Complex at Harwell (RCaH) in April 2010. The OCTOPUS (Optics Combined to OutPut Unique Solutions) cluster is a large scale imaging facility offered by the Functional Biosystems Imaging Group (FBI) of the LSF, which begun user operations and collaborative work in July 2010. OCTOPUS is a new concept in multi-modal imaging. Currently in Phase I of its development, the OCTOPUS cluster has at its core a hub of 14 lasers (3 Ti:Sa, 2 OPOs, 3 supercontinuum and 6 CW) (Fig. 1,2).

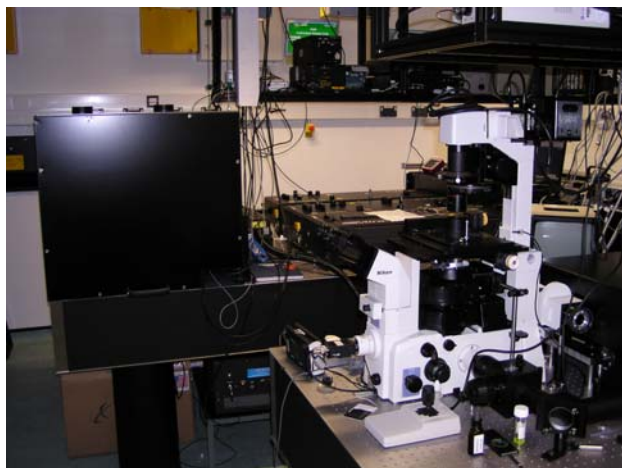


Figure 1. The laser hub. Left: The large black box holds the CW lasers and launchers (See Fig. 2). Right: the two Ti:Sa lasers (femtosecond and picosecond pulses) and an OPO. Bottom right: The two-photon confocal microscope housed in the laser hub.

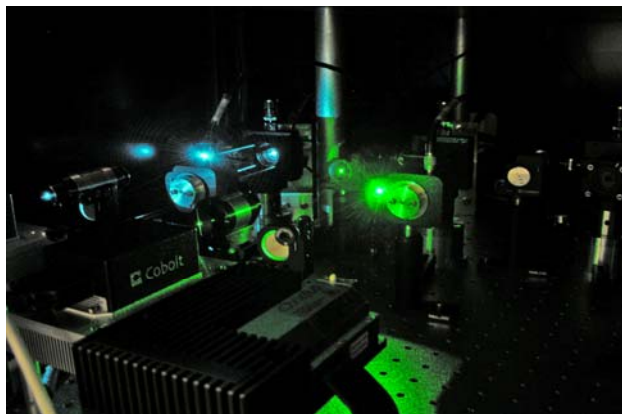


Figure 2. Two CW lasers launched inside the black box on the left of Fig. 1 above.

The laser sources provide excitation light to seven microscope systems, namely a total internal reflection fluorescence (TIRF) tweezers system, 3 single molecule fluorescence microscopes, each specialising in a different single molecule imaging technique, two 1-photon confocal FLIM systems and a 2-photon confocal FLIM system laid out as shown in Fig.3.

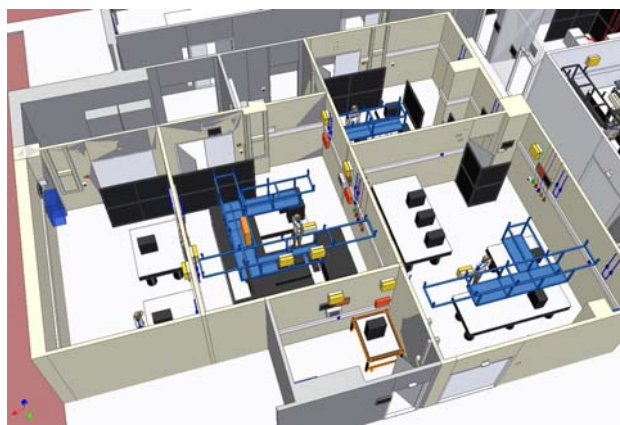


Figure 3. Layout of the OCTOPUS cluster. The central room is the laser hub with the different microscopes lab position around it. It also houses a two-photon confocal FLIM system. Bottom right: this room houses three systems, namely a TIRF-tweezers system, a one-photon confocal FLIM system and a 5-colour single molecule tracking system. Top right: This room houses a one-photon multicolour confocal FLIM system. Top middle: The control room (see Fig. 2). Top left: Storage room. Left: This room houses a 3-colour single molecule tracking system (top) and an 8-channel single molecule FRET and polarisation combined system (bottom). Bottom middle: Development room.



Figure 4. OCTOPUS laser control room. For more information contact Andrew.Tylee@stfc.ac.uk.

In Octopus different laser lines from the central core can be combined to supply 'engineered' illumination (i.e. with different wavelengths and different pulse repetition and length) which is tailor-made to the needs of the experiment. This is achieved through a combination of single mode optical fibres, fibre combiners, the AOTF's outputs from the supercontinuum laser sources and the outputs from the Ti:Sa and OPO sources. The fibres transporting light into the microscopes are interlocked through a central control room that keeps track of which laser is used in which microscope and ensure safe operation (**Fig. 4**). The first generation of OCTOPUS offers the following microscopy stations:

2 colour, 2 polarisation 8-channel single molecule TIRF microscopy. This is a home-built objective-type TIRF microscope illuminated via a single-mode fibre (**Fig. 5**). The excitation polarisation is purified using a Glan-Taylor polariser and then rotated between orthogonal polarisations using an achromatic liquid crystal polarisation rotator. The fluorescence is split into orthogonal emission polarisations in each of two wavelength bands, giving four channels which are imaged onto the CCD simultaneously for each of the two possible excitation wavelengths. Polarisation-sensitive dichroic mirrors are used to ensure that the fluorescence polarisation is not affected. The switching of the rotator is synchronized to acquire images such that alternate frames in a series have the same excitation polarisation. The combination of two colours and two polarisations allows us to monitor the distances between single molecules using Fluorescence Resonance Energy Transfer (FRET), as well as the orientation of the molecules using single molecule polarisation measurements. The microscope uses TIRF illumination, which restricts fluorescence excitation to a layer a few hundred nm thick to reduce the level of out of focus background fluorescence and to allow single molecules to be detected. Quad-View optics split the fluorescence into four spatially identical images on the CCD. The microscope can be used to image single fluorescent molecules in live cells, allowing the monitoring of molecular interactions in real time.



Figure 5. Left: Experimental setup for 3-colour single molecule tracking system. Right: experimental setup for multidimensional single molecule fluorescence imaging. The picture shows a wall-mounted black panel between the microscopes below the open yellow box on the wall. This black box houses the array of fibre connectors that allow laser light to be transported via fibres, combiners etc. from the adjacent laser hub in the OCTOPUS core to the microscope room through the wall. These boxes can

be found in all the labs. A fibre connector and cable is shown hanging below the black box.

3-colour and 5-colour single molecule TIRF microscope. This microscope is similar to the instrument described above, but allows the imaging of three different fluorophores simultaneously (**Fig. 3**). Multicolour microscopes are required for the investigation of macromolecular interactions in complex networks, for example signalling pathways. A five colour microscope is currently under commissioning (**Fig. 4**) (funded through a BBSRC Long and Large grant awarded jointly to STFC and King's College London).

Confocal Microscopy. OCTOPUS offers three confocal microscopy systems. The Multiphoton FLIM imaging capability located within the laser hub allows the imaging of molecules deep inside living cells (**Fig. 1**). This system is used in several collaborative programmes involving STFC and a number of universities.



Figure 6. Left hand side: one-photon confocal FLIM system. Right hand side: 5-colour single molecule fluorescence microscope.

A one-photon confocal FLIM system is currently under commissioning using a newly purchased Becker and Hickl scanning head (**Fig. 6**).



Figure 7. One-photon multicolour confocal system. In-house built confocal microscope that uses a Fianium supercontinuum as laser input.

OCTOPUS also has a multicolour confocal microscope used to characterise in situ the 3-D structural disposition of macro-molecular complexes at the plasma membrane of cells (**Fig. 7**). Both one-photon confocal FLIM systems have recently been upgraded with the addition of supercontinuum laser sources, which offers almost

limitless flexibility in the choice of fluorescent probes that can be used. Fluorescence correlation imaging has recently been commissioned on this microscope.

TIRF-Tweezers. The TIRF Tweezers setup is designed to combine the optical manipulation of samples with super high-resolution real-time TIRF imaging. A 1064nm laser is propagated through an AOD to enable a fully steerable trap which is focused onto the imaging plane. A Nikon TIRF cassette enables easy manipulation of a fibre laser beam past the critical angle of total internal reflection. The fluorescence emitted is imaged through a Nikon TE-2000 microscope onto a back illuminated Andor EMCCD camera. This will enable real time manipulation and force analysis of structures in biological specimens with minimal cell damage. There is capacity to upgrade the system to incorporate a super-continuum fibre input for multiple excitation and splitting optics for multiple-colour imaging as used in the systems above.

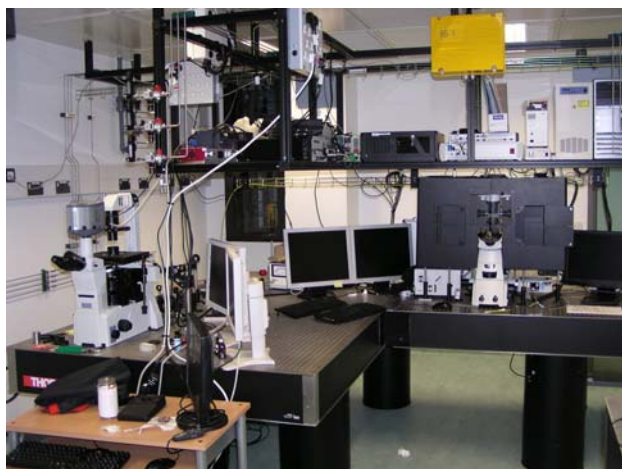


Figure 8. Left: TIRF-Tweezers set up. Right: One-photon confocal.

Phase I of the OCTOPUS cluster is now operational in the RCaH. The new location has allowed the integration of all the microscopy stations and laser sources, offering a fully flexible, large scale imaging facility to the user community.

Image Analysis. The analysis of data from single molecule measurements, particularly in live cells, is challenging because the images often have high backgrounds, signals are weak, and the molecules are mobile. The FBI Group has an active programme of development in advanced image analysis methods, using Bayesian techniques. This programme has been funded through a series of research council grants and the software is made available for use by the user community as it is developed.

The Future

Additional microscopy stations will be added as techniques and technology develop, and as the requirements of the community evolve. The location of the Molecular Structural Dynamics arm of the new LSF within RCaH will also allow the formation of an imaging “supercluster”, in which large scale pulsed laser systems will be used to image specific molecules in cells without the requirement for labelling.

OCTOPUS will also interact with other facilities on the campus, particularly Diamond Light Source and the proposed Imaging Solutions Centre. Proposed programmes include, for example, the combination of fluorescence, electron, and x-ray microscopy on the same sample, supported by information from macromolecular

crystallography and molecular dynamics simulations. This powerful combination, unique to the Harwell campus, will allow us to obtain a complete picture of how the structure of biological macromolecules enables them to fulfil their specific roles in the complex systems that are responsible for the functioning of cells and organisms in health and disease.

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New data analysis software for time-resolved spectroscopy experiments in the ULTRA laser facility

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Introduction

The ULTRA laser facility is a highly sensitive, time resolved spectrometer that provides valuable information on the structure and dynamics of molecules in solution. A diverse range of spectroscopy experiments can be performed within the ULTRA facility through its synchronised, dual (ps and fs) 10 kHz repetition rate output, with a broad range of wavelength tuning (from UV to mid infrared) [1].

ULTRA produces experimental data through the detection of spectral changes of laser light after transmission through a sample (see Figure 1). Scientists need to analyse this large collection of spectra within minutes to decide if their experiment is following their predictions or whether a modification to the experimental strategy is needed. The need for quick data analysis is vital in experiments where samples have a limited lifetime, potentially being light or temperature sensitive.

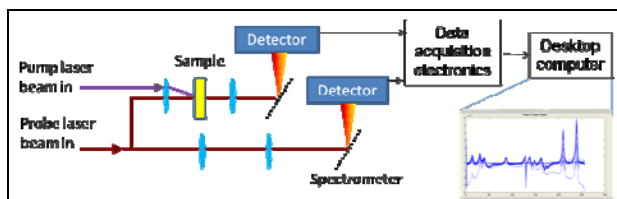


Figure 1 showing overview of typical ULTRA experiment for measurement of laser light absorption by sample. Note the use of two detectors, one immediately after the sample and the other as a reference for later data analysis.

The flexibility of the ULTRA facility introduces a number of variables that must be corrected for in order for meaningful data to be produced, such as variability in laser intensity between pulses, background intensity signals and electronic noise within detectors. The data analysis software was developed to remove these variations and is intended for use throughout the experiment at the Central Laser Facility and as 'take-away' software in the scientists' home institutions.

Software considerations

Previous data analysis performed using Excel spreadsheets was found to be time-consuming and laborious, especially with the increased number of variables and large volumes of data produced within the ULTRA laser facility. The new software automates this preliminary analysis process and presents the original and processed data in order to verify results, in order to confirm that the analysis has not introduced spurious effects in the data.

A set of functions were prioritised for development, principally to display spectral data and correct for instrument variation. The software was built in three stages (data collection and sorting, preconditioning and data display) and assumed the name 'Legobrick' software. A range of software tools were considered (including Labview, C++ and Matlab) and it was decided that Matlab offered the necessary range of functions (for data display, analysis and spreadsheet-like operations) and was familiar to the software developers (D. A. Robinson and M. R. Pollard). The software produces output files in the comma separated variable format (csv), a popular format that allows data to be accessed by other software such as Excel, Origin and Igor during development.

Data collection and sorting

This first elementary step requires the user to identify and load the data files containing spectral data. In the software, the data is assembled as an array, with axes attributed to experimental parameters (e.g. pump – probe pulse delay, polarisation, wavelength etc.).

Preconditioning

The second step removed the variation in data due to instrumentation effects and is shown in Figure 2. The preconditioning window displayed the experimental parameters in a spreadsheet-style data table with plots showing corrected data in two and three dimensions. Instructions for use of the software are given in the caption corner (bottom left in Figure 2).

The various functions corrected effects introduced by the apparatus or elements within the experiment. The instrument effects were corrected using two functions:

- Automatic identification and removal of values from data caused by 'dead' detector pixels.
- Overlap correction, where the spectrum is dispersed over multiple detectors for improved resolution and final spectrum is assembled from multiple detector data sets.

The following experimental effects were corrected:

- Separation of steady state and transient data, using the 'T0 Correction' function to select a value from the data table that represented the steady state background spectrum.
- Subtraction of reference spectral data with 'Reference processing', using data from a second detector (see Figure 1) that measures the intensity and spectral fluctuations of the laser.
- Correction for large scale difference spectra intensity variations using 'Baseline correction function'. This fitted a polynomial to user-defined points in the spectrum and subtracted the baseline from the dataset.

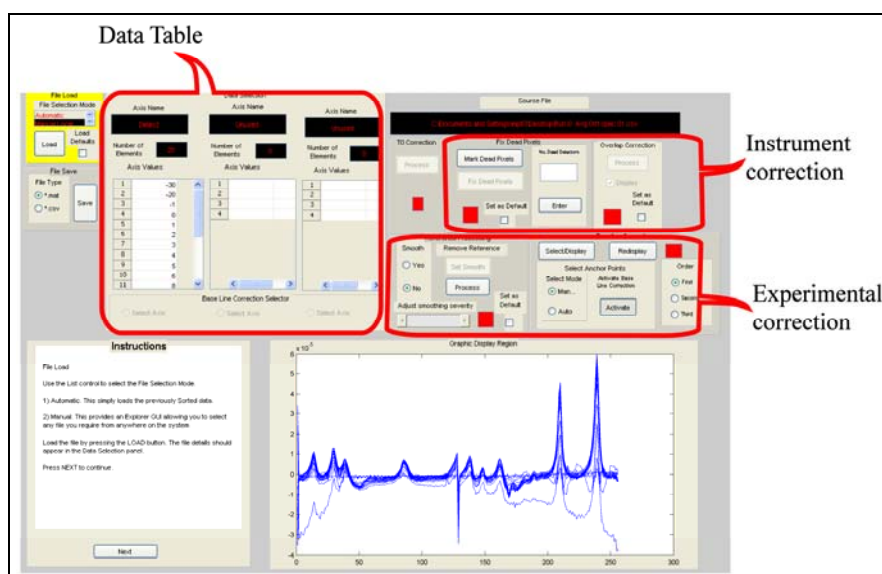


Figure 2 Screenshot of preconditioning window, with functions for data table, instrument and experimental correction highlighted in red

Graphics display

The final part of the software in this report allowed multiple spectra to be compared as one experimental parameter was changed. The graphics display window (see Figure 3) displays two and three dimensional plots of the spectral data, where a dimension refers to the experimental parameter. The parameters were displayed again in a data table format, which allowed users to plot data corresponding to an individual value or a combination of values, for example transient analysis of particular wavelengths or pixels (known as 'kinetics plotting'). The variation of the whole data set could be visualised using the 3d plotting function. A function called 'Release graph' allowed separate figures to be produced for use in journal publications.

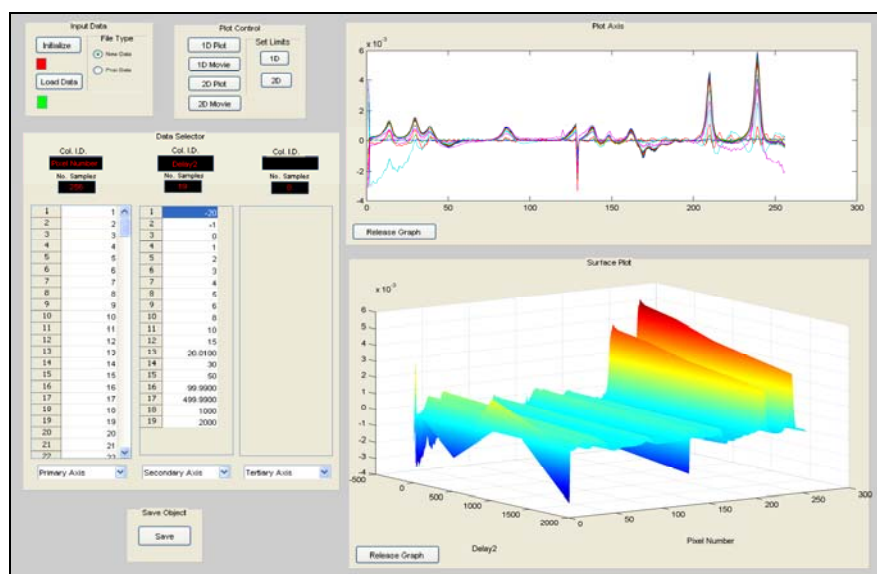


Figure 3 Screenshot of graphics display, showing data from sample of DMABN.

Conclusions

- The finished software is now in use in the ULTRA facility, with a typical day of analysis involving 15 experimental data sets, each consisting > 20 spectra. This permits scientists to efficiently make preliminary analyses and to inform the ongoing experimental strategy.
- The data analysis software has been well received by the user community and is used on site and in users' home institutions on a variety of desktop computers and laptops.
- Future versions of the software will allow for different experimental data sets to be imported for comparison and provide a calibration of the spectra relative to wavenumber.

Acknowledgments

We wish to thank D. A. Robinson for his efforts on this valuable work, despite a long period of serious ill health.

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Active synchronisation of dual amplifier outputs for time-resolved spectroscopy.

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Introduction

The ULTRA laser provides unique capabilities for advanced time-resolved spectroscopy techniques (such as Two-dimensional IR¹ and Femtosecond Stimulated Raman Spectroscopy²) that use the combination of dual 2 ps and 50 fs output pulses. The fs and ps amplifiers are synchronised naturally due to their common seed source but thermal drift can occur in the optical paths through the amplifiers and may result in relative timing drifts of several ps at the sample to be studied. Loss of synchronisation at the ps level can significantly affect measurements and so must be corrected. We report a control system that measures the relative timing between the ps and fs pulses and controls their synchronisation to a programmable fs value.

Relative timing measurement

The relative timing measurement takes a $\sim 1\%$ split of the 800 nm light produced from each of the two amplifier stages and cross-correlates the signals to extract timing differences.

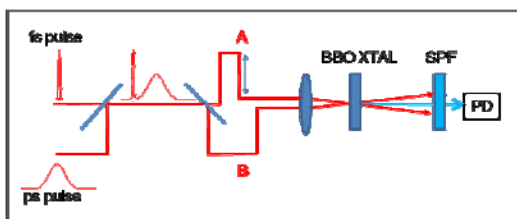


Figure 1. Optical apparatus for relative timing measurement between the ps and fs pulses.

The timing measurement apparatus (shown in Figure 1) combines the ps and fs pulses with a timing difference of ~ 10 ps. The combined pulses pass through a scanning cross-correlator consisting of two beam paths, A and B, with a variable delay on A. The pulses are recombined within a 1 mm BBO crystal (BBO XTAL), where up-conversion to 400 nm occurs when the pulses are overlapped in time. The up-converted signal is filtered by a short pass filter (SPF) and detected by a GaP photodiode (PD) whose output is sampled using a data acquisition card (National Instruments). The variable delay scans the relative timing of the pulses to produce cross- and auto-correlation peaks, as shown in Figure 2.

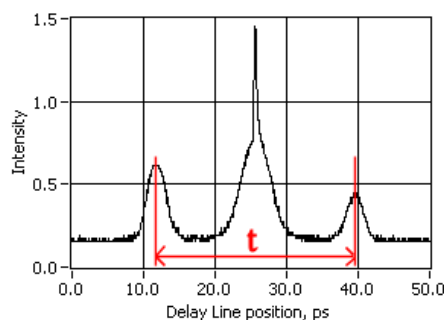


Figure 2. Waveform occurring from the correlation of ps and fs pulses in time.

The correlation waveform in Figure 2 shows three distinct peaks. The centre peak represents the combination of two auto-correlation signals and has a ~ 4 ps broad peak (mixing of ps pulse of arm A with ps pulse of arm B) with a narrow < 100 fs spike (mixing of the fs pulse of arm A with the fs pulse of arm B) at its maximum. The outer peaks in Figure 2 are ~ 2 ps broad and arise from cross-correlation of the ps pulse on one path with the fs pulse on the other path. For example, the ps pulse of arm A of the correlator (Figure 1) mixes with the fs pulse on arm B. The separation between these two cross-correlation peaks (shown as 't' in Figure 2) is equal to twice the relative timing separation between the ps and fs input pulses. The information from the waveform was processed using peak fitting and statistical functions written in Labview software.

Control system

A feedback loop (shown in Figure 3) was used to observe the relative timing measurement and correct any deviation from a defined set-point by adjusting the timing of the ps output pulses, with all functions performed in Labview software.

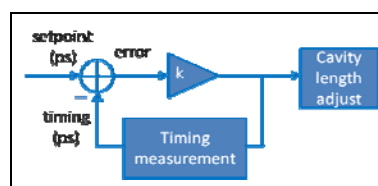


Figure 3 Diagram of control system using a feedback loop.

The timing of the ps output pulses was modified by adjusting the cavity length of the ps regenerative amplifier, by movement of one end mirror on a piezoelectric translation stage, with the magnitude of the adjustment controlled by the proportional gain value k (see figure 3).

Results

The relative timing measurement was performed over 2 hours and was found to drift by 1.5 ps. The control system was activated with a k value of 0.4 and controlled the relative timing to 42 fs rms, with a response time of 1 s, see Figure 4.

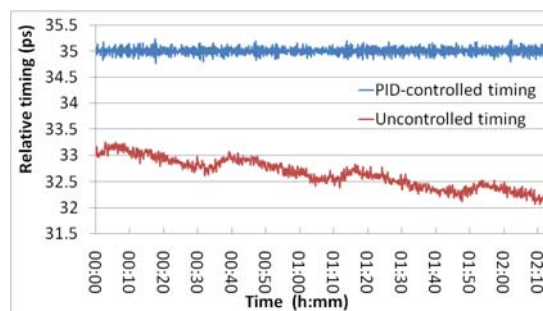


Figure 4 Comparison of relative timing over 2 hours.

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Optical trapping of laser targets under vacuum for ion-beam production in the LIBRA programme

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Introduction

The studies detailed here form part of the EPSRC Basic Technology programme (EP/E035728/1) entitled Laser Induced Beams of Radiation and their Applications (LIBRA). The concept of LIBRA is to develop a new generation of laser-driven ion-beam sources by using a high power laser beam to irradiate small plastic or liquid targets at high repetition rates. The laser's energy causes intense high-energy ionising radiation to be ejected from the surface (Figure 1). The ion-beam technology has the potential for wide ranging applications from cancer treatment and forensics to critical fault diagnosis and three dimensional chemical and structural analysis.

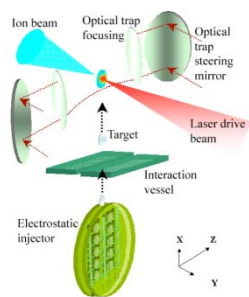


Figure 1: Original design concept for target delivery, optical trapping and target irradiation to produce ion-beams

LIBRA requires technological developments regarding targets, delivery systems, interaction environments, and detectors. The targetry and delivery requirements are foremost in this report as we intend to deliver targets to an interaction area under vacuum and suspend them without physical means of attachment. Physical supports and tethers are known to perturb the ion-beam production mechanism¹ and to generate unnecessary debris. We report the use of laser-based optical levitation and manipulation methods to achieve this goal.

Our aim is to levitate micron-scale targets, under vacuum, using photonic forces and to position the targets with micron accuracy into the beam path of a high power laser for ion-beam production. Here we demonstrate the underpinning methodology and optical requirements for trapping micron-sized droplets at low pressures in a vacuum chamber environment.

Laser Trapping Configuration

Two co-axially aligned, counter-propagating, laser beams (1064 nm) were used to create an optical trap through a balance of opposed scattering forces (Figure 2). Long-working distance objective lenses (working distance 10 mm) with high numerical aperture ($NA = 0.7$) were used to provide a robust and stable trapping environment for micron-scale targets. The working distance is sufficient to enable debris shielding and propagation

of ion-beams without interference from the optical components. The concept of counter propagation has been widely used in literature^{2,3,4} to trap aerosol droplets and we would like to develop this methodology further to enable its implementation inside a target interaction chamber.

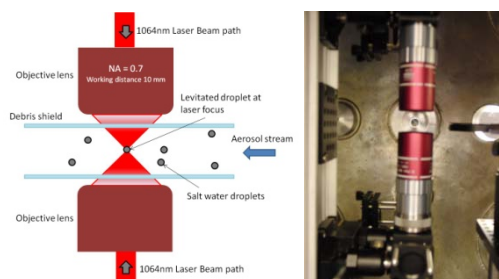


Figure 2: (left) Schematic diagram of the opposed focussed laser beams for optical levitation. (right) The centre point of the chamber is used to align the reference trapping position. The interaction laser enters through the port shown to the rear of the chamber.

For the optical trapping apparatus separate laser beams were required for each of the counter-propagating pathways. Therefore, two optical fibers were used to couple the laser into the optical path of the objective lenses. The use of fiber optic delivery was advantageous in enabling each of the counter-propagating pathways to be moved independently without realignment. The lower objective lens was mounted on an x,y,z stage to permit accurate alignment (to 1 micron). The laser power ratios were balanced using a combination of a neutral density wheel and the overall laser power. A steering mirror mounted in a conjugate focal plane to the back aperture of the objective lenses was used to adjust the laser focus positions and to create optimal trapping alignment. The trapped target and laser focus positions were imaged at x100 magnification to observe droplet stability (Figure 3).

Vacuum Chamber Modifications

The optical arrangement described above was constructed inside the vacuum chamber located in Laboratory 5A, R1 (see Figure 3). Several modifications were required to implement this set-up. The fiber optic cables were passed through the chamber walls using vacuum feed-through ports. The optical images were relayed by lenses through a chamber window to two CCD cameras external to the chamber and a motorized x,y,z stage was computer controlled from outside the chamber.

Experimental

In these initial experiments nebulised droplets of salt water, 1 to 5 microns diameter, were delivered in a flow stream to the trapping region whilst the chamber was open. A single droplet was trapped and the chamber was then carefully sealed and

evacuated. The evacuation was performed using a scroll pump which was pumping on a closed valve at the time of trapping. The valve was then carefully opened to start the evacuation. The procedure was used to prevent large vibrations, that occurred during the first few strokes of the pump when starting from cold, dislodging the droplet from the optical trap.

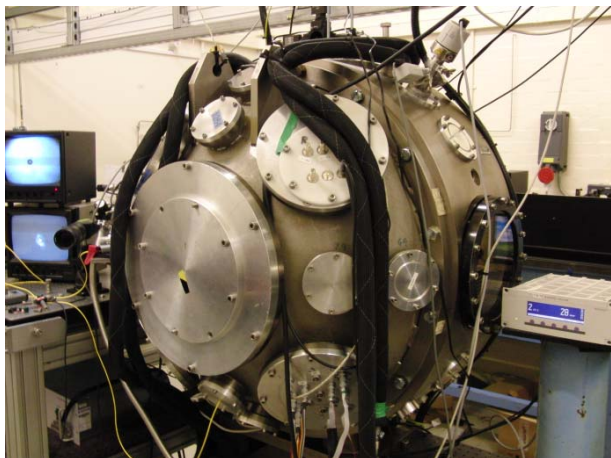


Figure 3: The vacuum chamber and diagnostics in operation. Droplet stability, laser alignment and chamber pressure are continually monitored. Trapping can be achieved from 1000 mbar to 5 mbar before efflorescence of the salt droplet occurs (i.e. a crystal is formed). The droplet is observed to be stable in position to approximately 1 micron.

As the pressure was reduced the position of the laser focus and the droplet were continually monitored (Figure 4). It was observed that the focal depth of the laser from the objective lens decreased by 52 microns as the pressure was reduced to below 20 mbar. The change was corrected using the motorized stage by decreasing the separation between the objective lenses. The droplet could be held at pressures of 9 mbar with a sodium chloride solution droplet and 5 mbar with a lithium chloride solution droplet. Beyond these pressures the droplet effloresced to a solid particle and was lost.

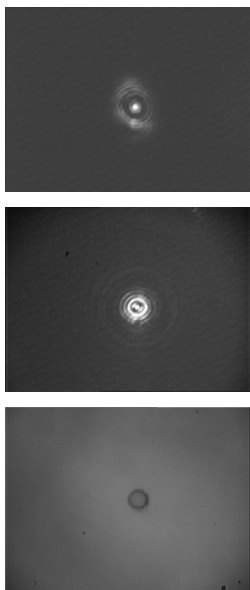


Figure 4: Images of; a) the laser focus from the lower objective, b) the laser focus from the top objective, and c) the levitated droplet. The droplet is 4 microns diameter and images are relayed from the top objective lens to a CCD camera.

Loading the trap was found to be robust and particles took less than a second to stabilise. There were no adverse effects experienced from pump vibration after the precautions outlined above. In addition, by gradual reduction of the chamber pressure no problems were experienced from air flows dislodging the droplet from the trap. From analysis of the images the target was maintained in position to within 1 micron during evacuation of the chamber.

Future Work

The next steps for extending this technology is to deliver solid particles and targets to the trapping region under vacuum conditions and to irradiate the target with a high power laser source to produce an ion beam. Solid targets will allow trapping at a higher vacuum and we plan to release micron-size particulates from dry surfaces using piezo-driven standing waves or to directly inject aerosolised solid particles.

Conclusions

We have successfully held a microscopic liquid droplet in the centre of a vacuum interaction chamber at 5 mbar using focused laser light to levitate the droplet. The droplet was held to a precision of 1 micron. By extension of this technology we aim to hold solid targets for the purpose of generating ion-beams under the LIBRA programme.

Acknowledgements

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Close-in Contrast Measurements of the New ps OPCPA Front End

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Introduction

In high intensity laser matter interactions the contrast of the laser pulse can be crucial to the outcome of the physics. In this article we discuss the addition of a picoseconds optical parametric chirped pulse amplification (OPCPA) stage to the Vulcan front end improving the amplified stimulated emission (ASE) by more than 2 orders of magnitude.

The technique of Optical parametric chirped pulse amplification (OPCPA) is becoming increasingly popular as pre-amplifiers for seeding laser systems [1,2] and as potential amplifiers for extremely high peak power systems. This is because they are capable of amplifying a broad bandwidth and can have large gains. Our previous study of the ASE contrast of the Vulcan laser system [3] demonstrated that the parametric fluorescence (PF) generated by the nanosecond OPCPA pre-amplifier acted to seed the rest of the amplification chain.

Picosecond OPCPA Preamplifier

The upgraded front end introduces a single picosecond stretch and then an OPCPA stage to generate a clean high energy seed pulse that is then injected into the Vulcan laser. Unlike other schemes [1], only the seed is stretched. This has a number of advantages that are explained below. Figure 2 shows a schematic of how this is achieved. The output from the Ti:Sapphire oscillator is divided into two with pulses being amplified in a Nd:YLF regenerative amplifier and stretched in a grating stretcher. As the pulses are amplified in the regenerative amplifier gain narrowing occurs that acts to increase their pulse length to 10ps. The output pulses are frequency doubled to 527nm with energies of 700μJ. These form the pump pulses for the picosecond OPCPA. The temporal profile of the output pulse means that the instantaneous small signal gain also varies. Consequently the seed pulses must be stretched to best match the pump pulse profile to extract energy efficiently and ensure close to uniform gain across the spectrum. The pump pulses are imaged into the BBO crystal and demagnified to a beam diameter of 1mm. The seed pulses are stretched to 3ps in a single grating stretcher. The stretcher is comprised of a grating with a line density of 1500 l/mm and an imaging lens as shown in figure 2. A retro-prism is employed to remove the spatial chirp and ensure a uniform beam. The stretcher has the added advantage that the lens and the back mirror are mounted on a single translation stage enabling the pulse length to be tuned. Since the picosecond OPA is degenerate a small ~2mrad non-collinear geometry is employed so that the seed and idler beams can be separated. The picosecond OPA generates pulses that have 70μJ of energy representing a conversion efficiency of 20% and are then injected into the existing nanosecond amplification system.

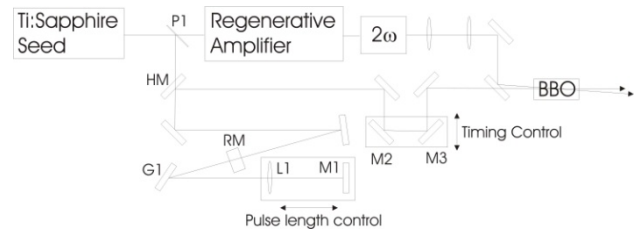


Figure 1: Schematic of picosecond OPCPA preamplifier

Rod Chain and Disk Amplifier Bypass

To examine the close in contrast to ascertain any changes introduced by the initial stretcher stage we have installed a beam line that by-passes the amplifier chain and relays the pulses from the front-end directly to the compressor chamber. This enables the use of a scanning 3rd order device to characterize the contrast. Figure 2 shows part of the bypass installed next to the main six in LA2 and LA3. Vertical propagation was enclosed.

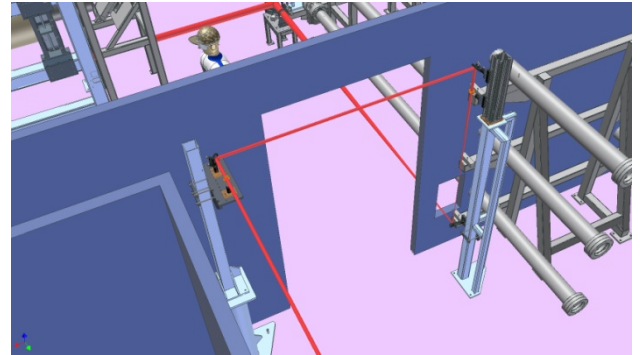


Figure 2: CAD drawing of part of the bypass in LA2 and LA3

TAP Diagnostics Line

The Contrast Monitor and 3rd order cross correlator (Sequoia) were set up on a diagnostics table outside the TAP vacuum chamber. There are several porthole windows that allow beams to be delivered out of the vacuum chamber. The beam must propagate several meters to the diagnostics table, maintaining spatial beam quality and size. The input aperture of the Sequoia is 5 mm and the beam must enter the device without clipping. Image relaying the beam using long focal length lenses ensures the spatial quality of the beam. The beam must be reduced in size by a factor of four in two stages to avoid breakdown of the beam in the air. Figure 2 shows the diagnostics beam line, set up to demagnify and image relay the beam out of the TAP vacuum chamber and on to the diagnostics table. After the compressor, the beam is 600 mm in diameter and focused on to the target centre via an f3 off axis parabola inside the target

chamber. The beam is recollimated using a 37.5 mm lens, mounted on the target stage. The target stage is motorized, allowing adjustment of the collimation after the gate value is opened under vacuum. The beam is reoriented using two, and it is reduced using a 2:1 telescope which also relays the beam through the window. The beam is collimated as it propagates through the window to avoid picking up aberrations such as astigmatism. The beam is image relayed through a second 2:1 telescope and arrives at the diagnostic table with a diameter of approximately 4 mm.

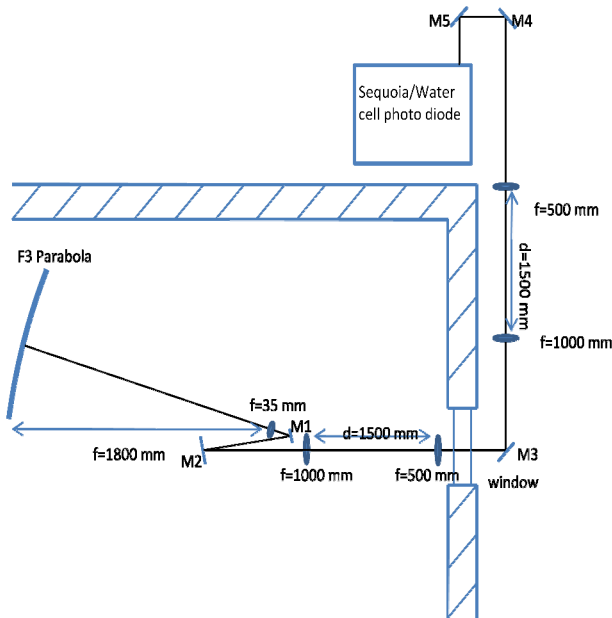


Figure 2: Layout of the TAP diagnostics line

Initially a single shot autocorrelator was used to adjust the ns stretcher to ensure that the 3 ps stretch had been compensated for. This confirmed that the pulse length was 500fs after compression.

Contrast Monitor

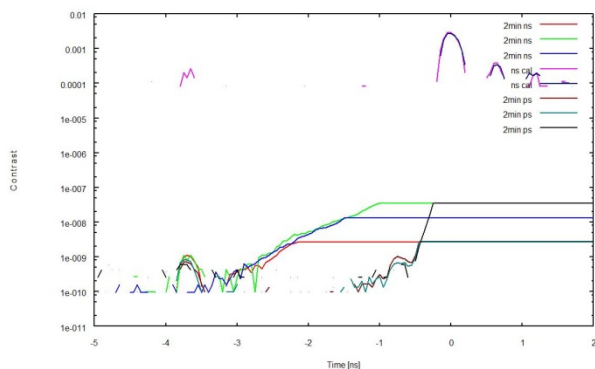


Figure 3: Contrast diode traces

A contrast monitor is a device that has been developed to measure the contrast of high energy, short pulses using a fast diode and a water cell [4]. Figure 3 shows a comparison of the photo-diode traces for the high and low energy seed input configurations. These traces were recorded using only the rod amplifiers and the OPCAs this is because the damage threshold of the collection optics restricts the amount of energy that can be generated in the laser system. The rod amplifiers act as spectral filters for the PF generated by the OPCAs limiting the PF's bandwidth to that supported by the rest of the amplifiers in the system. The disk amplifiers have small SSGs (Small Signal Gain) in comparison to the other amplifiers in the system so do not contribute significantly to the ASE level. As can be seen in figure 3 the dynamic range of the diode set-up is

10^{10} . For the low energy seed the ASE starts approximately 3ns ahead of the main pulse. For the high energy seed the ASE starts within 1ns of the peak of the pulse. The dynamic range for the diodes is achieved using an intermediate focus in a water cell to prevent damage to the diode and by using convolution to calculate the under measurement of the peak of the pulse.

3rd Order Cross Correlation

A Sequoia is a commercially available 3rd order cross correlator which is used to measure close in contrast, pulse pedestals and prepulses. It is a multi shot device that temporally scans the pulse. As can be seen from figure 4 the contrast drops to 10^8 at 30ps and corresponds to the dynamic range of the instrument, limited by the scatter or THG on the 2ω arm of the device. The trace shows that there are no significant pre-pulses generated by the front-end within 100ps of the pulse which equates to the scanning range of the instrument. Scans beyond the peak of the pulse also do not show any significant post-pulses that might become pre-pulses during the amplification process due to accumulated non-linear effects.

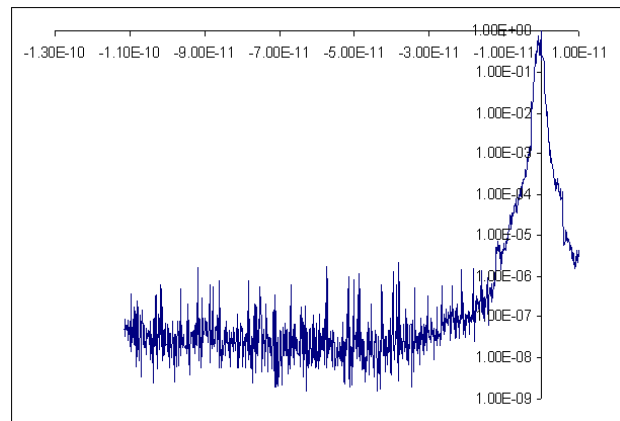


Figure 4: Sequoia temporal scan

Conclusions

In this article we have reported on the development of a novel dual OPCPA pre-amplifier system to seed high energy laser systems. That is sufficiently stable to be injected into the rest of the Vulcan laser system and leads to a direct improvement of the ASE contrast by at least 2 orders of magnitude without degrading the performance of the laser system. The Vulcan Petawatt laser system now has an ASE contrast of 10^{10} at 1ns, at 100ps the contrast is better than 10^8 and the PF caused by the ps OPA starts at 30ps. Further improvements could be achieved by increasing the amount of ps OPCPA pre-amplification, this might be achieved by adding an extra stage of amplification on the ps pump laser.

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Vulcan Rod Amplifier Upgrade

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Introduction

The Quantel rod amplifier control system and power supplies have been operating and running successfully on the Vulcan Laser System since the system was inaugurated in 1975.

We report on the programme to replace this control system and capacitor banks.

Reasons for replacement

The original suppliers of the equipment, Quantel SA [1], no longer manufacture or even support the electronics for the rod amplifiers and consequently the equipment has become obsolete. Therefore, it was necessary for the rod amplifier capacitors and control system to be replaced, as it posed a serious risk towards the future operation of Vulcan.



Figure 1 – Old Quantel units inside the Vulcan Main Control Room.



Figure 2 – Old Quantel rod amplifier racks inside Laser Area 1 that charged the 16mm and 25mm amplifiers.

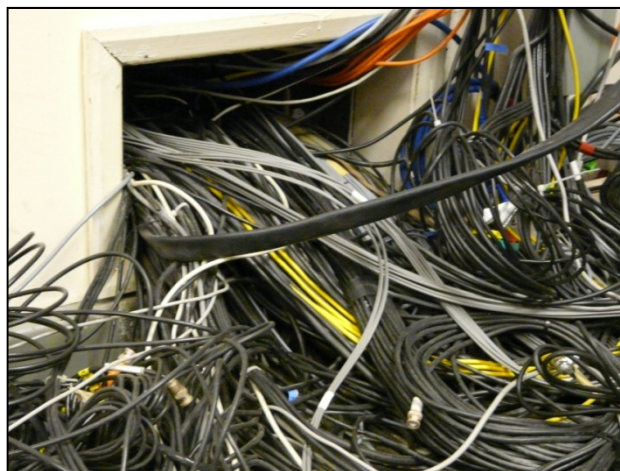


Figure 3. Cables running from the control room to the laser areas.

Figures 1 and 2 show the old Quantel units located in the control room and within the laser area. Figure 3 shows the cluster of cables coming from the only cable ducting in the control room to the laser areas - clearly unmanaged and with little capacity remaining.

Purchasing and preliminary testing

In order to minimize risks, the decision was taken to replace only the electronics (control system, power supplies and capacitor banks), not the amplifier heads since these were designed and built at RAL and could be maintained. Following a competitive tendering process the contract was placed with Quantel SA for replacement of capacitor banks and the hardware was purchased in 2008.

Testing of the new power supplies and control software was performed off line in 2008 proving the successful operation of individual rod amplifiers. An entire rod chain was powered successfully on the Vulcan laser system in April 2009. Operating the rod amplifier chain with the new capacitor banks showed an average (from 10+ shots) of 86.1% $\pm$ 0.4% of the old system (which is run with all amplifiers at 2.2kV). The maximum voltage of the new capacitors is 2.0kV. Operating the complete beamline at this lowered voltage provides sufficient system gain whilst extending component lifetimes.

Shutdown schedule

The Vulcan laser was shut down for three months from 12th August until 9th November to allow the replacement of the rod amplifier racks from inside the control room and laser areas with six new racks inside Laser Area 1. The timetable was as follows :

- Weeks 1-7 : Old equipment removal, cable labeling and rationalization
- Weeks 8-9 : New Quantel equipment installed in Laser Area 1
- Week 10 : Commissioning new power supplies
- Weeks 11-13 : Re-commissioning Vulcan – beam alignment and timing

Removal of equipment and cabling

Optics inside the laser areas were sheeted down to prevent accidental damage and dust accumulating on their surfaces. There was no direct access to remove racks from the middle of Laser Area 1. A vacuum spatial filter was removed and many low-lying cables were disconnected to allow the old Quantel racks to be moved out of the area; this was a common theme during the project in order to manoeuvre the large cumbersome racks.

The shutdown period was also an opportunity to clean up the Vulcan laser area and control room in general, as an accumulation of cables had built up over the last 30 years. This has led to an increased difficulty when installing new systems when many old systems have not been completely removed from the area, taking up unnecessary space and causing even more confusion in the area.

A total of 400 cables were labeled and documented, and then disconnected in order to facilitate the removal of the existing Quantel power supplies. The control desk was also removed and some of the redundant equipment was donated to Imperial College with documentation. Strict cable management was vital to complete the project on time. This involved keeping an up to date record of all the cables that were disconnected, removed redundant cables, tidying cable runs and salvaging the Quantel rod amplifier cables from the system.

The unmanaged cable situation had built up over the long operating lifetime of Vulcan. The result of improved cable management is that whereas it was previously unmanaged and confusing, tracking systems of cables is now far more streamlined enabling the installation of new hardware.

Installation of new equipment and system commissioning

By mid-October the new power supplies had been installed. A significant visual improvement was achieved with more walkway space available on both sides of the racks (Figures 4 and 5).



Figure 4 – Laser Area 1 with new Quantel hardware installed.

Many redundant systems have been removed, reducing confusion amongst staff and mess in Vulcan. Approximately a thousand cables were disturbed during the shutdown. If one of these were a working cable in the system and was not correctly reconnected the system would not function properly. A few trigger cables for cameras were found to be not reconnected properly during the start-up, which were quickly diagnosed and fixed.

Software issues became apparent during testing, which were eventually resolved. The new rod amplifier electronics were commissioned with very few problems, and the project overran

by only three days. The first shot after the shutdown achieved the required energy with the new Quantel rod amplifier racks.



Figure 5 – New Quantel racks, located on north wall of Laser Area 1, to power the 45mm rod amplifiers and control processing and power of both rod chains.

Refurbishment of the main control room

The Vulcan Main Control Room is intended to become the focal point for visitors to the CLF. Once the new Quantel racks were installed, a new space had become available in the room for a larger Vulcan controls and diagnostics desk; allowing more monitors, PCs, communications and working space. Along with this new operations desk, new display furniture has been installed for improved operations. In addition, the new control room allows for more effective communications during external visits to the CLF.

Conclusion

The upgrade was successfully completed three days overdue and user experiments commenced on Monday 9th November 2009.

Although the project as a whole was a success, there were potential pitfalls that could have caused the project to fall far behind schedule, such as lack of staff co-ordination and available expertise. The experience gained from this project will aid the future success of the upcoming six-year 10 Petawatt upgrade project.

Reference

1. Quantel SA, 2 Bis Avenue du Pacifique BP23 91 941 Les Ulis cedex, France. www.quantel-laser.com.

Vulcan Computer Control System Upgrade

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Introduction

Apart from the obvious need for significant numbers of laser optics and components, the successful operation of the Vulcan high-power laser over the past several decades has been heavily dependent upon sophisticated electronics, high voltage pulsed-power units and computerized control systems. This paper reports recent major changes to the computer control systems.

Historical Background

The computer controls on Vulcan originated (in the late 1970's) with a GEC 4080 "mini" computer but in the mid 1980's this was removed in favour of a pair of IBM 286 DOS-based PC's. Over the following decades the network gradually grew in size and complexity with (by the late 1990's) some peripheral PC's running Windows NT / 2000 but with the core fundamental laser control PC's remaining DOS.

As for the two rod amplifier chains, although these had seen a number of mechanical and optical re-designs, the essential pulsed power of capacitor banks, charging units and electronic control units have remained those as originally supplied from Quantel, France, over 30 years ago.

With loss of key people with their knowledge and experience as well as deteriorating serviceability of the respective software and hardware, both the main Vulcan computer and the Quantel control systems were ripe for replacement.

The Conversion

The project to rewrite the sophisticated computer control system, converting everything to windows XP, turned out to be more complex than originally thought and became a four year project. The bulk of the development for this took place offline with the final windows controls being tested during the normal inter-experimental maintenance periods, a process which ensured minimal operational downtime.

The new windows control software was installed in parallel with the existing DOS controls so that, as far as was possible, online testing could be facilitated and the control functions switched between the two systems fairly quickly.

The DOS was "switched off" in June 2009 and (after a successful operational period using the new system) was finally removed during the September - November shutdown.

The full laser control network now comprises of a dozen PCs running Windows XP and with around 80,000 lines of code (predominately Delphi and LabVIEW). The three main (and independent) touch-control screens are depicted in figure 1 below with (left) the disk amplifier / capacitor bank control screen; (middle) the main control screen with many control functions and diagnostic report forms and (right) a layout diagram showing the schematic of the laser.

In addition there are many diagnostic PC's which are sent various commands during operations to facilitate shot sequencing, data capture and archiving.

Conclusions

The system has been operational for the bulk of this past reporting period and has proved to provide a much more flexible and future-proofed operational system which has benefited both laser operations and the experimental user programme.

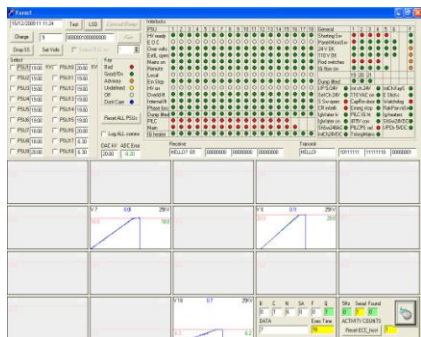


Figure 1a - Disk amplifier control and diagnostic screen showing capacitor bank interlocks and high-voltage charging graphs.



Figure 1b - Main control screen giving access to prime control functions and diagnostic data such as the shot firing sequence and waveform displays.

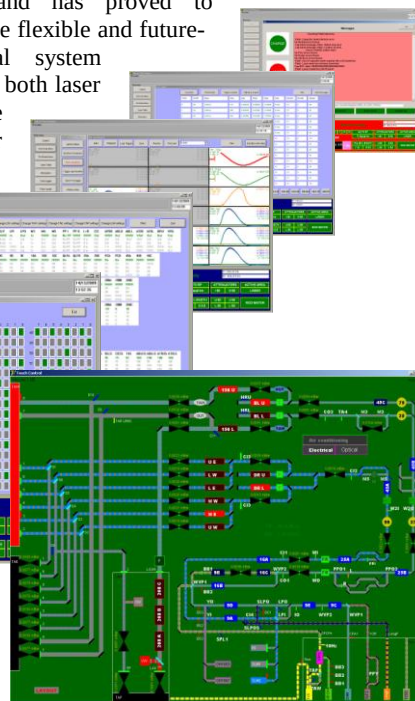


Figure 1c - Schematic of the laser providing direct control of components via the touch screen interface.

The Vulcan 10PW project – Building Design

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Introduction

A key aspect of the Vulcan 10 PW project is the new building that will house the large amount of equipment required for the 10 PW laser facility. A basic conceptual design was developed during phase one of this project¹⁾. This design extends the current building (R1) to maximise the available space within the limited surrounding land at RAL.

In preparation for the start of phase two of the project a team consisting of STFC staff and external consultants has been working hard for the past six months to develop the Vulcan 10PW building design.

During this period the design brief has been agreed, initial surveys conducted, risks reviewed, the layout developed to incorporate operational requirements, disabled access, fire strategies etc, and the structural design and services outlined. A value engineering exercise has also been conducted to prioritise the various design elements against the available budget.

The 10PW management team have now agreed a conceptual design for the building that has been costed to a level adequate for the project to proceed to the building detail design, ready for anticipated tender action in the Autumn of 2010. This article describes the agreed building design concept.

Building Objectives

The primary building objectives were:

- To provide a building that is fit for purpose in relation to the occupants and equipment that it will accommodate.
- To provide a building that caters for the installation, and possible future removal, of major items of equipment.
- To meet the financial constraints of the project. The current building project budget is £5m (excluding contingency).
- To meet the project timescales. The current 10PW project program schedules handover of the building in the Autumn of 2012 .
- To provide a design which can be built with minimal impact upon ongoing CLF (and other departments) activities in the existing and nearby buildings.

Overview of areas

The area that the 10PW building extension will occupy is shown by the red lines in figure 1 and by the yellow shaded area in figure 2. The building will comprise of:

- New HIA (High Intensity Area) combining current TAE (Target Area East) and South Control Room extended to the south.



Figure 1. Location for 10PW building extension.

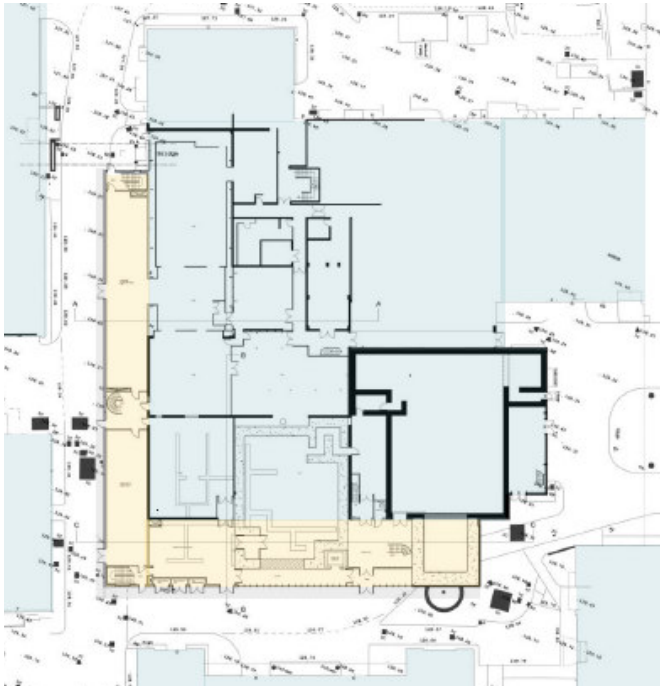


Figure 2: Site Plan showing extent of 10PW building extension

- New Target Area Control Room (combined for TAW & HIA) to the south of TAW (Target Area West).
- New Optical Parametric Chirped Pulse Amplification (OPCPA) Compressor and Diagnostics Area (Laser Area 7) above the current TAW & new HIA.
- New OPCPA long pulse amplification laser area (Laser Area LA6) to the west.
- New OPCPA front end laser area
- New OPCPA laser control room
- New laser amplification area (Laser Area 5) to the west for possible additional long pulse beams.
- An extension of TAP (Target Area Pettawatt) to the south with a shielded Electron Beam Dump Area.
- New Pulsed Power Area to the west.
- New entrance/atrium for the Vulcan facility.
- New building and CLF plant services areas.

Image

The design team developed a number of key words and phrases that attempt to encapsulate the image that the CLF wishes to portray in the design and detail of the facility:

- Open and interactive
- Modern and impressive facade
- Progressive
- Light and airy
- Excellence
- Safe and healthy
- Accessible to all
- Conscious of the environment

Primary Design Determinants

The primary design determinants are as follows:

The CLF attaches great importance to the design quality of all installations and this must be continued in the design and delivery of this facility.

The design proposals should provide opportunities to encourage interaction between staff.

Security arrangements must allow 24hour, 7 days per week access for designated staff and users. CCTV supervision for security of the new facility will not be incorporated however CCTV between control rooms and target/laser/ancillary areas is required for safety reasons.

Where possible the design should seek to utilise environmentally friendly, low carbon, sustainable solutions and energy efficient features. The use of natural daylight will be considered as well as the appropriate use of materials and M&E technologies.

Comfort, air quality and environmental stability for equipment and apparatus must achieve best practice for such an installation.

Design proposals will take full cognisance and support the implementation of CLF logistical and safety requirements.

Statutory regulations, including Building Regulations and the Disability Discrimination Act (DDA) require that proper provision be made for people with disabilities, though exemptions may be sought where the nature and severity of the disability is inherently incompatible with good health and safety practice. The new visitor access areas will comply with DDA.

Systems should be as simple as possible with adequate, safe, easy access provided to all parts of the installations. They should also be designed taking into account the limited resources available to the STFC for operating the plant. It is important that M&E systems are designed so that they can be repaired, maintained, inspected and extended with minimal disruption to the building user. Maintenance access to the rooftop of the new facility will be provided if necessary for servicing of building plant.

Viewing locations for high profile areas such as laser and target areas will need to be provided.

The functional areas will need to be located and linked in a cost effective and operationally practical and safe way.

The preferred strategy for moving major items of laser equipment into and out of the building is via the west elevation at each level and into the HIA from the South. The lifting and transportation of large heavy equipment can be achieved using specialist hired equipment and contractors and via temporary access routes if necessary. Fixed cranes will be provided within LA7, the HIA and the TAP electron beam dump area to move the equipment.

A lift will also be used for moving smaller items of equipment between floors.

There are a large number of below ground site services to the south and west of the current building and these will need to be moved/accommodated in the final design.

TAW will be temporarily closed during the build phase and then will be brought back operational again. To minimise the down time of the current Vulcan facility during the build phase, the CLF currently proposes not to strip out all the equipment from TAW.

The TAP area will not be operational during the build phase.

The current liquid Nitrogen tank will need to be relocated and the required capacity of the CLF nitrogen system will need to be assessed.

The design will be reviewed during the development stages to assess the impact upon CLF and other departments operations i.e. all CLF lasers particularly the Astra Gemini laser in the adjacent R7 building, SSTD in R25 and ISIS in R2 and R3.

The current HVAC, vacuum infrastructure, Nitrogen tanks and gases associated with TAW will need to be relocated/replaced.

The new facility will incorporate a new front entrance for staff, users and visitors. The front entrance should be welcoming and project the image that the CLF wishes to portray to the outside world. It should be light and airy.

The design life of the building, as defined in BS 7543, should be 60 years.

The components, assemblies, and installations, that are to be specified shall have a design life, as defined in BS 7543, of not less than 15/20 years.

All design will be undertaken in accordance with the Building Regulations and the appropriate British Standard Codes of Practice and other specific guidance notes relating to the particular installation.

Layouts

The ground, 1st and 2nd floor layout of the 10PW laser building are shown in more detail in figures 3, 4 and 5 respectively. The yellow shaded area indicates the extent of the new build.

Working from top to bottom and left to right of figure 3, the ground floor accommodates: space for additional long pulse beamlines, a capacitor bank, a TAW/HIA target area control room, the new HIA target area, an entrance area and the TAP electron beam dump area.

Likewise figure 4 shows the first floor which includes: main building air conditioning plant room, double height capacitor bank, target area services room, double height HIA, entrance landing and double height TAP electron beam dump area.

Figure 5 shows the second floor which houses: the OPCPA pump laser area, the OPCPA amplification and compression laser area, the OPCPA front end room, the 10PW laser control room and the laser area services room.

Finally figure 6 shows several elevations and cross sections of the building extension and figure 7 and artist's impression of how the building may look when completed.

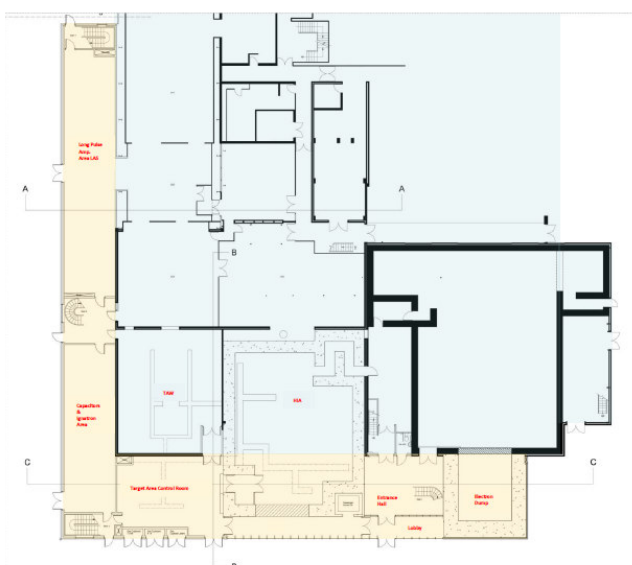


Figure 3: Ground Floor Plan

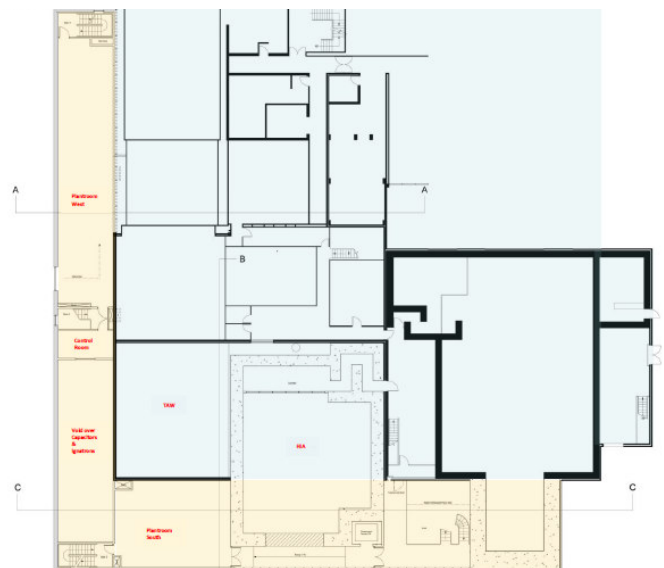


Figure 4: First Floor Plan

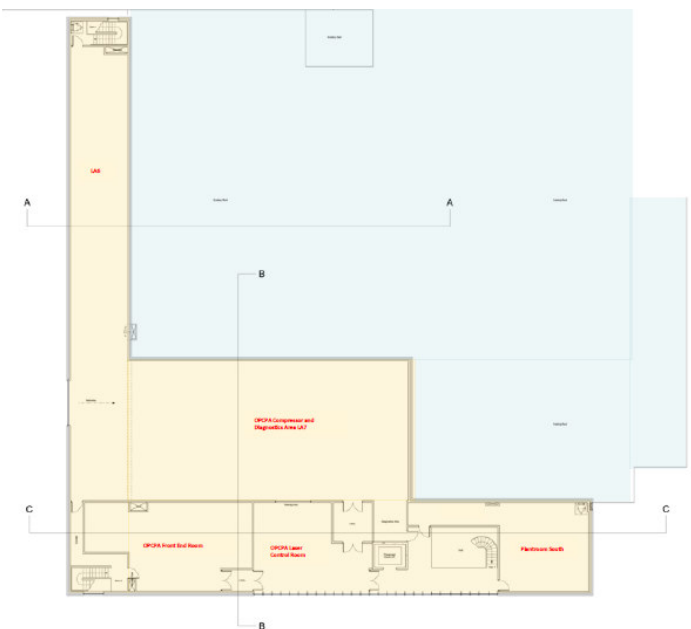


Figure 5: Second Floor Plan

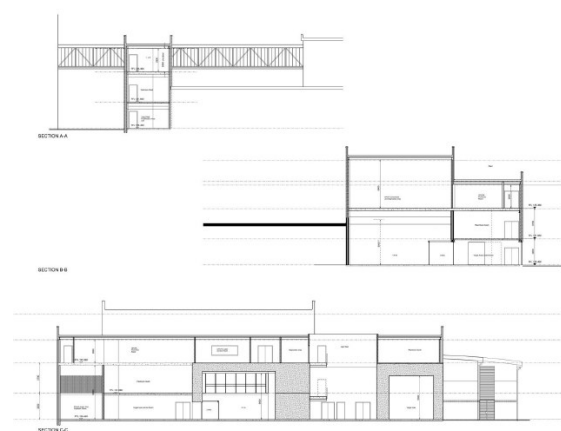


Figure 6: Elevations and sections



Figure 7: Artist Impression of building from South West corner

Programme

The proposed building project programme is shown below. The programme, and the procurement and contract strategy which it incorporates, are based upon the following assumptions:-

- Internal consultations required by the Client's organisation can be undertaken to comply with the timescales shown in the Programme.
- Client approvals can be provided to comply with the timescales shown in the programme.
- Design, manufacture, testing and commissioning of the new laser equipment will be progressing in parallel with delivery of the building.
- CLF will be able to provide sufficient details of its proposals for fit out of the building such that the interface with the building design can be co-ordinated during Stages 2 and 3.
- Funds will not be available for any site works to commence until the start of the 2011/2012 financial year.
- CLF operations within Building R1 will stop in 2011 such that site works can commence at the start of the 2011/2012 financial year.

The key dates for the project, as depicted by the programme, are:

Milestone :	Date:
Stage 2 Commencement	May 2010
Submit planning application	August 2010
Issue tender documents	November 2010
Contract award	April 2011
R1 Vacant Possession	April 2011
Site works commencement	May 2011
Site works completion	August 2012
Client Fit-out commencement	September 2012

Conclusions

This article has outlined the current design status of the 10 PW building.

Over the next few months much work will be ongoing to develop a more detailed design ready for tender action. A series of client review and value engineering stages will ensure that it will remain a functional, and affordable building which will meet the requirements of the laser and target area equipment to be installed in it and to ensure that it can be built to the project's timescales.

References

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The DiPOLE project: towards high energy, high repetition rate diode pumped lasers

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Introduction

DiPOLE stands for Diode Pumped Optical Laser for Experiments. It is a new project at the CLF to develop the foundations of novel high energy, high average power laser systems based on diode pumped solid state laser (DPSSL) technology. Compared to conventional systems, this approach promises dramatically increased repetition rates (and hence average powers) at significantly higher electrical-to-optical conversion efficiency. DiPOLE has been included as an emerging opportunity in the Research Councils UK Large Facilities Roadmap [1].

Motivation

Laser amplifiers capable of producing energetic ns-pulses are one of the main tools for laser plasma research and high-energy applications. Laser chains containing such amplifiers can produce ns-pulses or ps-pulses if the chirped pulse amplification (CPA) technique is used. Depending on the application, these pulses are either applied directly or are used to pump other amplifiers (e.g. Ti:Sapphire or OPCPA) in order to obtain even shorter pulses in the fs-regime.

Currently, ns-amplifiers are based on flashlamp-pumped Nd:glass technology and their repetition rate is limited to a few shots per minute for amplifiers delivering tens of joules of pulse energy to a few shots per day for lasers delivering kJ-level pulse energies.

Increasing the repetition rate of such laser systems to the multi-Hz level (typically 10 Hz) is pivotal for the following applications:

- Opening up new horizons in fundamental laser plasma interaction research by enabling higher throughput and the exploration of larger parameter spaces.
- So-called secondary sources which use laser-generated plasmas to produce ultra-short pulses of energetic particles (electrons or ions) or electromagnetic radiation (ranging from THz to hard X-ray). High repetition rate drive lasers are required to generate sufficiently high particle and photon numbers. Much of the pan-European ELI project focuses on the development and exploitation of secondary sources [2].
- Inertial confinement fusion (ICF), which is expected to be demonstrated for the first time within the next two years. Whereas current low-repetition rate facilities like NIF and LMJ are suitable for proof of principle experiments, high-efficiency, high repetition rate DPSSL based laser drivers open up the possibility to develop ICF into a reasonably clean, practically inexhaustible source of energy. This is the focus of the pan-European HiPER project [3].

Amplifier concept

The main activity within DiPOLE is the development of a DPSSL amplifier concept that is capable of delivering kJ-level pulses at 10 Hz repetition rate.

Gain material selection

The starting point was the identification of the most suitable gain material. This material needs to provide:

- A long fluorescence lifetime to minimise the number of pump diodes required
- Good thermo-mechanical properties to handle the high average power
- Reasonably high gain cross section to enable uncomplicated and efficient energy extraction
- The possibility to be manufactured in large sizes to handle the high pulse energy.

Ytterbium (Yb) as an active laser ion offers very long fluorescence lifetimes, a low quantum defect (pump wavelength 940 nm, laser wavelength 1030 nm), reasonable gain cross sections and efficient high power laser diodes are readily available for its pump wavelength. Two host materials have been identified that offer good thermo-mechanical properties and can be manufactured in large sizes: crystalline calcium fluoride (CAF) and ceramic YAG.

Yb:CAF has a very long fluorescence lifetime of 2.4 ms and a large gain bandwidth (> 50 nm) [4], and is therefore a promising candidate for directly diode pumped chirped pulse amplification (CPA) systems for producing sub-ps pulses [5]. However, since it exhibits a very small gain cross section, very large fluence levels are required for both pumping and extraction in order to achieve good optical-to-optical (o-o) efficiency.

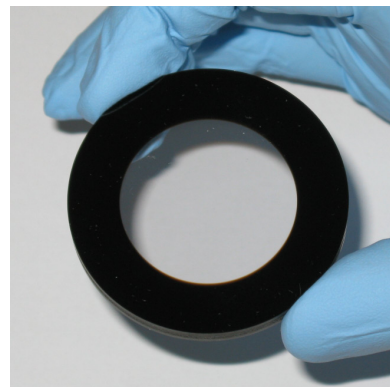


Fig. 1: Yb:YAG - Cr⁴⁺:YAG compound disk.

On the other hand, Yb:YAG has an order of magnitude higher gain cross section with a reasonable fluorescence lifetime of 1 ms [6]. Since the main application of our envisioned kJ-class laser is the production of ns-pulses, either for pumping amplifiers for fs-pulse generation (Ti:sapphire or OPCPA) or for driving inertial fusion targets, we think that ceramic Yb:YAG is the best choice for the gain medium. Also, monolithic compound structures with different doping species are possible with ceramic YAG [7]. The photo of such a

compound disk is shown in Fig. 1. Here the inner region of the disk is doped with Yb^{3+} and acts as the active laser medium, the outer region is doped with Cr^{4+} which heavily absorbs at 1030 nm and therefore acts as an index-matched absorber for suppression of amplified spontaneous emission (ASE) [8].

Efficiency and gain modelling

Numerical modelling has been carried out to determine optimum amplifier design parameters. In the model, the storage efficiency η_{stor} has been calculated for various parameters like pump fluence, pump pulse duration and pump spectral width. η_{stor} is defined as extractable fluence divided by pump fluence. Loss mechanisms and associated efficiency factors that influence η_{stor} are: η_{flu} for the fluorescence decay, η_{QD} for the quantum defect, η_{abs} for pump light that passes through the gain medium without being absorbed and, finally, η_{reabs} for the minimum upper state population that needs to be established in order to overcome reabsorption that exists due to the quasi-3-level nature of Yb:YAG. If a pump pulse duration of 1 ms is chosen, η_{QD} and η_{flu} limit η_{stor} to 58 %. It turns out that η_{abs} and η_{reabs} need to be balanced off against each other and that for a given set of pump-related parameters, there is one optimum value for gain medium optical depth (OD = thickness times doping concentration) that yields the maximum η_{stor} . If OD is chosen too low, too little pump light is absorbed, if OD is too high, reabsorption losses become dominant.

The following results are calculated, unless stated otherwise, for an amplifier that is end-pumped from both sides with a pump pulse duration of 1 ms, a 5 nm FWHM pump spectral width, centred at the optimum wavelength in the 940 nm absorption band. Spectrally resolved pump absorption cross sections were taken from [9]. Quantities calculated were η_{stor} and the small signal gain G , defined as $G = \exp(\eta_{\text{stor}} F_{\text{pump}}/F_{\text{sat}})$ where $\eta_{\text{stor}} F_{\text{pump}}$ is the extractable fluence and F_{sat} the gain saturation fluence. First, calculations were carried out for room temperature operation. The results are shown Fig. 2. It becomes apparent that very strong pumping is required, firstly to overcome the high reabsorption losses and to achieve good efficiency and secondly to overcome the still rather low gain cross section and achieve reasonable gain. The required high pump and extraction fluences are difficult to achieve because of limited pump source brightness and limited laser damage threshold.

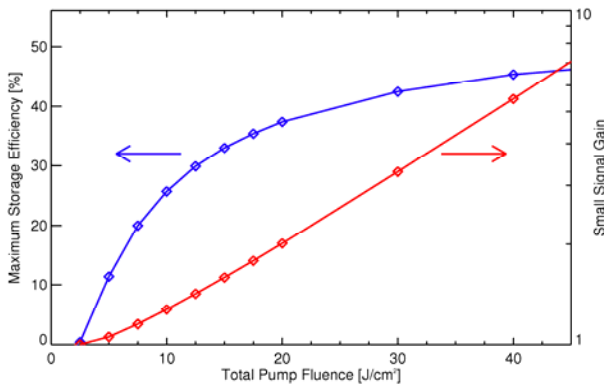


Fig. 2: Maximum storage efficiency (blue) and small signal gain (red) for amplifier operated at room temperature.

Cooling the gain medium to 175 K drastically changes the situation, as illustrated in Fig. 3. Reabsorption is reduced and the gain cross section increased, leading to greatly improved efficiency and gain, especially at moderate fluences. A pump fluence that is realistically achievable with today's laser diodes is 10 J/cm^2 (5 J/cm^2 from each side), yielding a storage efficiency of just over 50 % (resulting in an extractable fluence of 5 J/cm^2) and a small signal gain of 3.8. This fluence is

therefore chosen as the preliminary operating point for our amplifier.

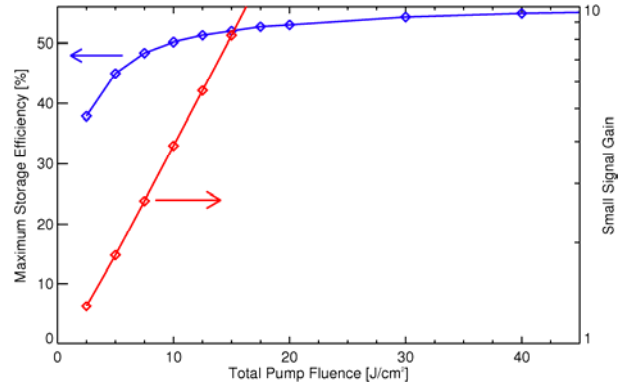


Fig. 3: Maximum storage efficiency (blue) and small signal gain (red) for amplifier operated at 175 K.

Operating at cryogenic temperatures yields the added benefit of improved thermo-mechanical and thermo-optical properties like increased thermal conductivity and reduced temperature dependence of the refractive index [10]. Another effect is the narrowing of spectral features both in the emission and the absorption spectrum. The absorption spectra measured at room temperature and at 175 K are shown Fig. 4, together with a 5 nm FWHM Gaussian spectrum for comparison, which is assumed to be the spectral shape of our pump source.

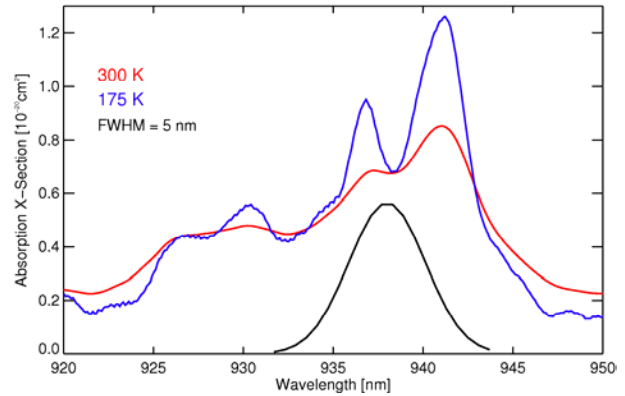


Fig. 4: Absorption spectra of Yb:YAG at different temperatures and 5 nm wide pump diode spectrum for comparison.

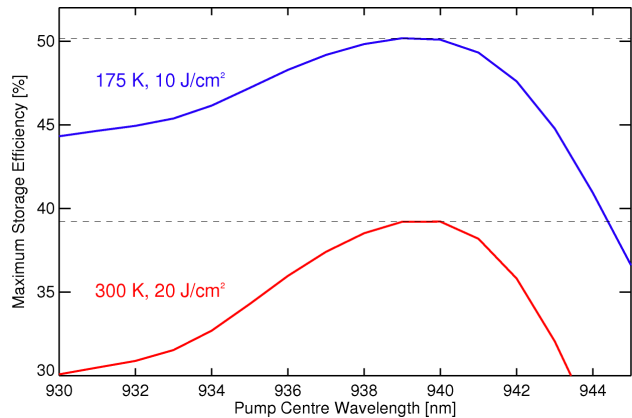


Fig. 5: Storage efficiency as function of pump centre wavelength for two different temperatures and pump fluences.

If these pump and absorption spectra are used to calculate storage efficiency for two different temperature scenarios, results as shown in Fig. 5 are obtained. Even though the pump fluence in the low temperature scenario is only half that of the room temperature case, significantly higher storage efficiency is predicted. Lower temperature operation also shows a much

weaker dependence on pump centre wavelength. So despite narrower absorption features, the requirements with respect to spectral performance of the pump diodes are less critical for low temperature operation.

Amplifier geometry

After determining operating temperature and pump fluence for our envisioned amplifier, the actual geometry needs to be defined. If the laser system is to yield an output energy of 1 kJ and the amplifier is to be operated at an output fluence of 5 J/cm², the aperture needs to be 200 cm² or 14 x 14 cm² if a square beam shape is adopted. The optimum OD = $N \times l$ obtained from the numerical calculations is 3.15 % cm, where N is the Yb-doping concentration in atomic % and l the geometrical thickness of the amplifier. The choice of N and consequently l is governed by ASE management considerations. If the gain-length product along the diagonal across the (square) surface of the amplifier is to be kept below 3, we require $N < 0.18$ % and hence $l > 18$ cm. Such a thick amplifier requires distributed cooling as demonstrated on the Mercury laser [11]. There, the gain medium is divided into a stack of thin slabs with He gas flowing through the gaps between the slabs. The concept is illustrated in Fig. 6, where an amplifier consisting of 10 slabs is shown.

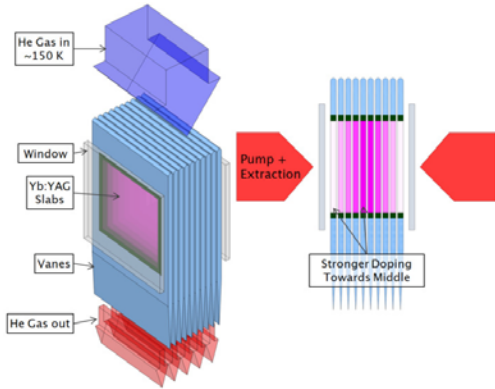


Fig. 6: Illustration of amplifier geometry: isometric (left) and side view (right).

If the criterion that the transverse gain-length product must not exceed a certain value is applied to each individual slab, one realises that the doping concentration can be increased towards the centre of the amplifier. The advantage is twofold: firstly, since the required overall OD remains the same, the amplifier as a whole becomes thinner, saving material and reducing the impact of nonlinear effects, and secondly the optical power absorbed in the individual slabs and hence the heat load is equalised. An optimised doping profile for a 10-slab amplifier is shown in Fig 7, together with the transverse gain as a function of position. The overall thickness of the gain medium is reduced from 18 cm to 10 cm in this configuration.

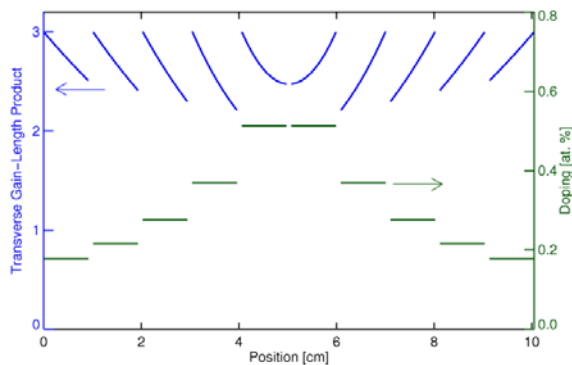


Fig. 7: Transverse gain-length product and doping levels along optical axis in 10-slab amplifier.

DiPOLE prototype

To test the concept in the laboratory a lower-energy multi-J prototype amplifier system is currently being built. The design is based on four co-sintered ceramic YAG discs (55 mm in diameter x 5 mm thick) where the Yb-doped region (35 mm diameter) is surrounded by a 10 mm thick Cr⁴⁺ cladding to absorb unwanted transverse fluorescence. A photograph of one of these discs is shown in Fig. 1. Given the 2 cm total gain medium thickness, two different Yb doping levels of 1.1 and 2.0 atomic% have been chosen to maximise storage efficiency and to equalise the heat loading. The discs are mounted in a vacuum insulated pressure vessel through which cryogenically cooled He gas is flowed. A schematic of the cryocooler system is shown in Fig. 8. The amplifier is end-pumped from both sides by two diode pump lasers operating near 940 nm. Each source provides 20 kW peak power in pulses of ~1 ms duration in a square beam (2 x 2 cm²), with a corresponding pump intensity of 5kW/cm² and at a repetition rate of 10 Hz.

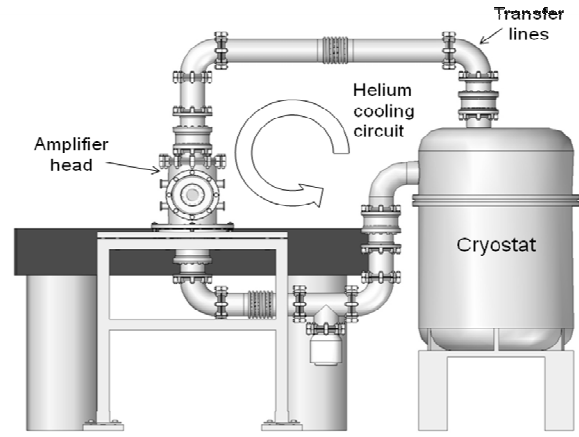


Fig. 8: Schematic diagram of the cryocooler system under development at CLF.

Conclusions

In summary, we have presented the conceptual design of a cryogenic Yb:YAG amplifier that can be scaled to kJ energy levels and beyond, owing to its geometry and unique cooling technique. Considerable enhancement in optical-to-optical conversion efficiency is predicted with the reduction of pump fluence at low temperatures owing to the reduced reabsorption losses, increased pump adsorption and gain cross sections. Numerical modelling also shows a weaker dependence on pump centre wavelength at low temperature, relaxing the stringent requirements of the diode pump source. The introduction of doping gradient in the slab architecture helps facilitate a uniform gain distribution and heat load in addition to the reduction of the overall thickness of the amplifier medium from 18 cm to 10 cm. A lower-energy multi-J prototype amplifier system is currently under development at CLF as a proof of principle.

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Timing and Synchronisation System Designs for the New Light Source

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Introduction

The New Light Source (NLS)^[1] was a proposed UK and international user facility based around a combination of Free Electron Lasers (FELs) and conventional lasers. In 2009 it was decided to halt work on the proposal once the Conceptual Design Report had been completed. Among the subsystems described in the CDR^[1] was a sophisticated laser-based timing and synchronisation network, designed by CLF staff.

The NLS was designed to support a very wide range of science, a significant fraction of which exploited its ultrashort pulse capabilities and, in particular, its ability to deliver several beams to an experiment simultaneously. The relative timing of the pulses in these beams had to be tightly managed, either to overlap them reliably in time, e.g. for multiphoton experiments, or to provide them with variable relative delay(s) e.g. for pump-probe studies. In addition to the lasers delivering beams directly to users there were also lasers acting as components of the NLS machine itself. The timing of the pulses from these needed to be controlled to ensure that the facility functioned at all. This article describes the systems used to manage the timing and considers the performance of an example subsystem in more detail.

Time Structure

The NLS time structure was decided in consultation with the scientific user community. The outcome was particularly simple, driven by a strong preference for the pulses from each FEL and laser to be delivered in a continuous, evenly-spaced, high-rate train. (Experiments needing a lower rate would have the option of pulse selection.) As far as the timing was concerned the only parameters to be decided were, therefore, the pulse duration and repetition rate. The duration was chosen to be 20 fs FWHM to allow the best possible time resolution. There was a clear science requirement for an initial rate of at least 1 kHz with an upgrade path that would increase this, in approximately decade steps, to 1 MHz. In principle exactly these rates could have been chosen. But in fact slightly different rates were selected with a view to minimising the timing jitter of the resulting pulses.

The starting point for the rate was the frequency of the RF modules which accelerated the electrons for the FELs. This was predetermined by industry standards to be 1.300 GHz. The NLS operating rate would need to be an integer sub-multiple of this and for technical reasons the timing jitter would be minimised if the integer chosen had only very small integer factors itself (ideally only 2 and 3). The eventual scheme is shown in Fig. 1. The overall master clock frequency was chosen to be 216.67 MHz, the sixth harmonic of which would generate the 1.3 GHz RF. The nominal 1 kHz pulse rate to be delivered on Day 1 of operations was actually 1.102 kHz (1.3 GHz divided by $2^{17} \times 3^2$). Subsequent upgrades would increase it by factors of 2^3 , $2^2 \times 3$ and 2^3 , eventually reaching 0.846 MHz. An additional feature of this choice was that the rate after each upgrade would be an integer multiple of the rate before. This would make it easier for legacy systems to remain operable after any upgrade.

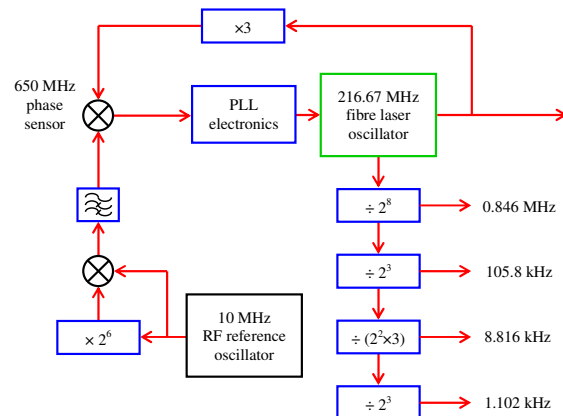


Figure 1. NLS master clock architecture showing pulse rate division scheme.

Synchronisation

The potential that ultrashort pulses offer for very high temporal resolution and also for high-intensity multiphoton science can only be achieved if the relative pulse timing is very tightly managed. For a subset of NLS experiments it would be sufficient if the timing management consisted simply of pulse-by-pulse timing measurement followed by time-stamping of the resulting data. However there were also experiments for which this approach would not be adequate and, in particular, there were aspects of the machine operation (for example the use of conventional lasers to seed the FELs) where the timing would need to be *controlled* rather than just measured. For this reason a synchronisation control system was designed which would be deployed across the whole of the NLS.

Given the 20 fs FWHM chosen pulse length, the target figure for *overall* synchronisation was set at 10 fs RMS. This was recognized as a challenge since individual subsystems have only been developed to this level of performance in recent years. However the NLS would have an advantage over almost all similar facilities in that it would be built from scratch without having to accommodate any pre-existing hardware or design decisions. This would allow the need for low timing jitter to be factored into the design from the start.

The entire synchronisation system is too complex to describe here. Instead the general approach will be illustrated by an example. One of the features of the NLS was the configuration of its FELs as seeded amplifiers with the seed pulses derived by high harmonic generation from conventional lasers. The timing of the FEL pulses would then be set by the timing of the seed lasers and the stability of the output beam transport paths. This scheme imposes two separate synchronisation requirements. Firstly that the seed pulses must overlap the electron bunches in the FEL undulator. The nature of electron accelerators, whose transport systems are inherently dispersive, makes this difficult to achieve. The fact that the NLS accelerator did achieve it^[2] is testament to the very considerable effort and creativity of the design team. But the bulk of that work concerned the accelerator itself rather than the synchronisation system and it will therefore not be discussed further here.

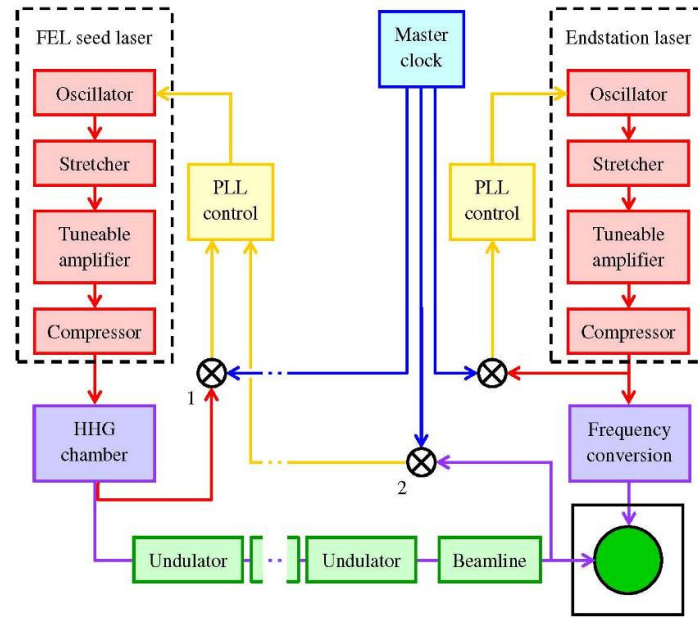


Figure 2. Layout of the elements used to synchronise FEL and laser pulses at the users' experiments. 1 and 2 represent the first and second phase sensors for the FEL seed laser.

The second challenge is the delivery of the FEL pulses to the users' experiments in exact synchronism with pulses from a conventional "endstation" laser. The scheme devised to do this is shown in Fig. 2. The physical scale of the system is large - the seed laser and the endstation laser are more than 100 m apart. Yet the need for less than 10 fs timing jitter corresponds to a relative movement of the pulses in space of less than 3 μm . So issues around the transport of beams and of timing signals are critical. For this reason a master clock architecture was chosen (the details of the low-noise clock are shown in Fig.1). The alternative would have been to use a common laser oscillator and to split the beam from this to feed the two amplifier chains. The first problem with this is that transporting ultrashort pulses over long distances is difficult. To avoid nonlinear effects the transport has to be in free-space (ideally in vacuum) using curved mirrors for image-relaying to preserve the beam quality. Engineering such a system for high stability would be very expensive. On the other hand the technology needed to transport optical clock pulses with few-femtosecond jitter has already been developed and demonstrated^[3].

The second problem with a common oscillator is that the active control system needed to correct the pulse timing in real time ideally uses the position of an oscillator cavity mirror as its actuator. Such a mirror is multi-passed very many times on the timescale of the correction signal (typically $>10 \mu\text{s}$). So the mirror need only be moved a tiny distance, which eases high speed operation. Of course if there was only one oscillator then movement of its mirrors would not correct the *relative* timing of the endstation laser and FEL pulses. So an extra-cavity actuator would have to be used which would be less effective.

Having distributed accurate clock signals to the phase (timing) sensors in Fig. 2, the next task is to reduce the timing error they detect using a closed phase-locked loop (PLL). Again this is a well-established technique and locking of a laser oscillator to an external reference using optical phase sensing has also been demonstrated at the femtosecond level^[4]. Extending this technique to include the laser amplifier chain inside the timing control loop should be straightforward, the only complication being the reduction in pulse rate relative to the oscillator. This imposes a Nyquist limit on the phase noise spectrum which can be sensed. However given that the lasers are designed to operate at up to 1 MHz it seems likely that this limit will be higher than the one imposed by the correction actuators.

Once the seed laser is phase-locked FEL action can begin and the opportunity then exists to correct timing jitter arising in the FEL and beam transport paths. This will involve sensing the pulse timing after the FEL, at point 2 in Fig. 2. This will be considerably more challenging than sensing the laser timing a) because phase sensing in the EUV, where the FEL operates, is not yet a mature technology, b) because the FEL will be tuned over a much wider photon energy range than the laser and c) because the uncorrected noise levels (both amplitude and phase) from the FEL will be higher than for the laser systems alone. The effectiveness of the correction system depends directly on these levels. However despite these problems a post-FEL correction scheme has been included in the design as it promises the easiest route to reaching the overall jitter target. The tuning issue will also apply to the endstation laser if it proves necessary to stabilise its timing after its nonlinear frequency conversion. The initial plan is to achieve stability passively, by mounting the converters close to the endstation and in a well-controlled environment. But the option of active stabilisation has not been discounted.

Conclusions and Acknowledgement

Some elements of the timing and synchronisation scheme for the NLS have been described. The timing is expected to be straightforward. Delivering 10 fs RMS overall synchronism will be challenging, but it should be achievable using state of the art subsystems. This work was partly supported by "IRUVX-PP", an EU co-funded project under FP7 (Grant Agreement 211285).

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Experimental setup in the Vulcan HaPPIE Laboratory for Multi-beam Combination to achieve Diffraction limit pulses

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Introduction

High powered lasers are attractive owing to their potential for a diverse range of experiments. In order to achieve these high powered lasers, various choices for amplification of laser beam have to be made. These choices include the size of the beam which in turn requires laser crystals or ceramics to be manufactured close to their current limit of scalability. Another way of achieving these high power requirements is to combine several beams into a monolithic beam, which immediately reduces the requirements for the amplifier to a more modest level. This requires technology to lock the beams spatially and temporally.

A number of projects are underway in Europe under the umbrella of the LASERLAB-EUROPE II to research high power laser beams. There are several initiatives under this known as Joint Research Areas (JRA) of which one of them is the **High Average and Peak Power lasers for Interaction Experiments (HAPPIE)** to target the technology areas which are deemed useful to achieve high power lasers. One of these technology areas is the control of the spatial and temporal overlap of combining these beams in a coherent manner.

Specifically in the case of laser fusion project, known as HiPER¹ it is envisaged to include a fast ignition beam. This fast pulse ignition beam requires ~60 KJ with Picoseconds pulse width. This beam is to be composed of multiple subbeams in order to achieve the required power levels for fast ignition. A number of these subbeams will be combined to act as monolithic beam. This also reduces the requirements on the power from each individual amplifier stage and gives a level of control on the different subbeams for pulse shaping. In order for the laser power to achieve the diffraction limit at the interaction point, it requires that the subbeams be temporally and spatial overlapped. It is also currently thought that the main igniter beam would now also compose of subbeams.

Another pan European laser project called Extreme Light Interaction (ELI); which involves development and construction of three separate lasers in different geographical locations. It is currently thought that each laser will be constructed from several laser beams which also require spatial and temporal overlap.

Spatial overlapping can be achieved by several techniques²; most prominently this is achieved in the Large Telescopes either in use or currently being developed around the world. These large telescopes are constructed from segmented mirrors and the wavefront needs to be flattened to within a fraction of the wavelength to achieve the diffraction limit to fully exploit the capability of the telescope. We are currently setting up a laboratory to investigate the different techniques in wavefront measurement spatially and temporally.

Wavefront measurement methodologies

In the case of wavefront measurement there are several different methods which are employed. These methods traditional

measure the wavefront of one aperture and correct this wavefront. The two most common methods is a Shack-Hartmann wavefront sensor and an interferometer based technique via the multiwave lateral shearing interferometer⁴. A Shack-Hartmann technique samples the wavefront with the use of a grid of microlenses which is focused onto a sensor, generally a CCD camera. In the case of a distorted wavefront the focused points on the CCD camera are misaligned relative to an undistorted wavefront which is proportional to the distortion in the wavefront. The position of focused spots compared to a set of reference focused points for a "perfect" wavefront is used to create a phase map which can be extracted of the measured wavefront or a Fourier demodulation technique, which obtains an error signal. The phase map for one aperture is generally expressed as a set of either Zernike or Legendre polynomials. Error signal can then be used to correct the wavefront by the use of an adaptive mirror. Adaptive mirror contains actuators to control the shape of the mirror so that the distortion can be corrected in the wavefront. In the case a multiwave lateral shearing interferometer uses a 2D diffraction grating which replicates the incident beam into 4 identical waves which are propagated along slightly different directions. These directions differences create interference patterns. After a few millimeters propagation the four beams are slightly separated. The grid deformations are directly connected to the phase gradients. A spectral analysis using Fourier transforms allows the phase gradient extraction in two orthogonal planes. The phase map is finally obtained by integration of these gradients. As explained in the case of Shack-Hartmann sensor the phase map can be used as an error signal to an adaptive mirror. These methods do not provide a piston error, this being the amount of lateral movement one beam has to make with the second beam so that they are spatially overlapped as they propagate, so cannot be used to spatially overlap two beams.

To create a phase difference between two independent sub apertures then it is important to interfere the two wavefronts. This then creates an interference pattern between the two wavefronts from the sub apertures and then the phase information (piston measurement) can be extracted. In order to obtain this piston measurement between two sub aperture beams then a quadriwave lateral shearing interferometer (QWLSI)⁴ can be employed. This measurement is self referenced, which makes it easy to implement. This method adapts the well known multiwave lateral shearing interferometry used for wavefront sensing to phase difference evaluation by extending the shearing distance so that the edge of one subbeam overlaps the edge of another subbeam. Analysing the interference pattern in this overlap region leads to this phase difference between the two subbeams. One particular system designed to do this is the PHASICS camera SID4. This was developed in collaboration with the CEA CESTA who developed the system for the phasing of two LIL compressed beams⁵. The piston accuracy can be estimated to be 40 nm or less.

If we consider two beams with complex amplitude $A(B) = \sqrt{I_{A(B)}} \exp(j(\psi_{A(B)})) + c.c.$ then split the beam into a 4 wave interferometry. We get a case of independent subbeams, and to obtain the simplest beamlet combination, it is necessary to align the physical gap direction between the subbeams with the interferometer replication grating in axis y. In this case we obtain a situation similar to the 2-wave case. We observe 4-wave interferences, with the two wave interference zones at its left and right. The 4-wave interference zone labelled OR is illuminated by four replicas, A1, A3, B2 and B4. In the OR zone, the pattern results from the amplitude double product $(A_1 B_2^* + A_3 B_4^*)$. The argument of this sum leads to a phase difference that is equal to $[(\psi_{A1} + \psi_{A3}) - (\psi_{B2} + \psi_{B4})]/2 \approx \psi_A - \psi_B$. In the left (right) zone, the pattern results from the interference of replicas from the same initial beam A (B). The argument of the corresponding amplitude sum leads to $A(B)$ beam phase gradient. Considering an unaberrated beam, this phase gradient should be equal to zero or to a constant if the wavefront is tilted. In the case of unaberrated beams a graph of the total amplitude argument versus x may represent a top hat shape where the amplitude, $P(x)$, represents the phase difference between the two independent subbeams A and B. Furthermore, in the case of angularly phased subbeams, its amplitude would be equal to the piston difference between them. A deeper theoretical study shows that a deconvoluted signal in the overlap zone allows characterization of not only the piston but also tip and tilt and higher order aberrations.

Current Progress

Under the JRA there is a specific task that requires the development of novel metrology and techniques to lock a number of pulses of discrete test large apertures together through active phase control. These test apertures are of a small size so that any technical difficulties can be solved at this point in so that they can be scaled to a larger size required for current planned facilities. With this in mind at CLF we have developed a laboratory to assess various techniques for beam phasing experiments. The initial experiments will be conducted in CW mode of a Ti:Sapphire laser output. A PHASICS camera is employed for capture and analysis of the combined wavefront. As shown in Figure 1 the optical layout expands the beam to a 120 mm beam from the laser, so that it can be subdivided into at maximum four beams of 50mm square. Although initially the experiment will start by using two beams of 50 mm square each. The two mirrors have independent control of tip / tilt and a translation stage for each mirror for piston control. Mirror 1 is the slave to Mirror 2 so that when a piston error is calculated by the PHASICS camera and software, this translation stage is aligned to bring the piston error close to zero. In Figure 1 the combined beam is relay imaged from the two square mirrors onto the PHASICS camera, to ensure that the plane of reference is imaged onto the focal plane array of the camera. The beam condenser system composes of a 570 mm lens and a 100 mm lens to collimate the beam. This makes the beam slightly larger than the aperture (4.8 mm by 3.8 mm) of the camera. Wavelength range of the sensor is 350 - 1100 nm with a spatial resolution of 29.6 μm . There is also a beamsplitter so that the far field image of the beam so that the diffraction limit of the spot is measured. The PHASICS camera is connected to a laptop to analyze the wavefronts and in the future implement the phase correction. Figure 2 actual shows the arrangement in the laboratory to date.

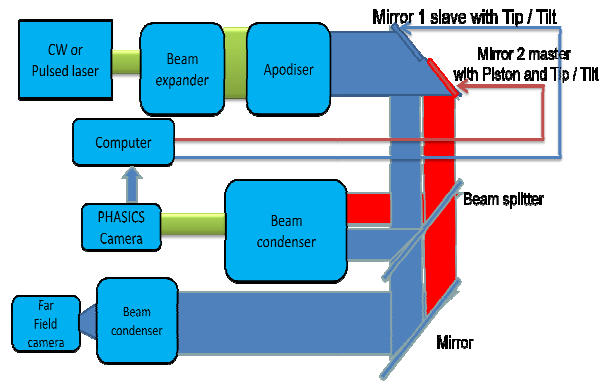


Figure 1. Experimental layout.

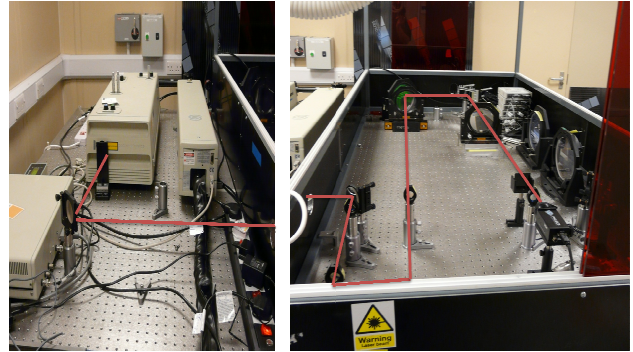


Figure 2. Laboratory setup, beam path is shown by the use of the red line.

The current PHASICS software on the laptop displays the two subbeam wavefront measurement plus interference between the two subbeams. The interference area can be adjusted by varying distance between the grating and the CCD sensor up to a maximum. The distance is defined by the beam parameters and the distance that separates the two beams. An algorithm is then employed to calculate the difference in phase between the two beams, which is displayed as a fraction of the wavelength being measured. This is carried out by the use of Legendre polynomials as opposed to Zernike polynomials due to the fact that the wavefronts are square. The wavefronts can be measured with the use of Zernike polynomials by defining the area as circular in the software. The piston and discrete Tip / Tilt errors does not yet go to the translation stage and their respective stages. With the purchase of a programming language from PHASICS, which will give us access to the analyzing software and then it will be possible to introduce a feedback loop to control the piston error and the discrete Tip / Tilt, which is what we are planning to do in the immediate future.

This consists of a Ti:Sapphire Tsunami working from 0.8 to 1.1 μm , 100 fs pulses at 80 MHz pulse rep rate.

Future Developments

This project is funded by the LASERLAB-EUROPE II fund.

We will develop the system for analyzing the wavefront for a feedback control system, so that the two beams will be locked spatial. This will initially be carried out on a two beam system, which will be expanded to a four beam system. This will require further mirrors and a different configuration to arrange the mirrors. We also require further development of the software so that three of the mirrors receive error signals to spatial overlap them.

We would like to implement a basic interferometer system so that an estimation of the distance between n numbers of mirrors can be calculated in the first instance. In which the PHASICS

camera can then finely adjust the n numbers of mirrors in the system.

Once the system is in a working state and fully tested in the HAPPIE laboratory then we would like to develop the system on the Astra-Gemini laser system here at CLF, to spatial overlap the two beams.

We also like to explore techniques to temporal lock several beams together.

Conclusions

We have a setup in the HAPPIE laboratory with the current technology to carry out analyses of subbeams wavefront to spatial phase them. In the future we are going to design a feedback system so that the two beams are permanently spatial locked after this we would implement the system on Astra-Gemini.

We are also going to explore experimentally the temporal locking two picoseconds pulses in the future.

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Installation of the Target Fabrication Quality Management System

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Introduction

With target complexity increasing and also with the introduction of high repetition rate laser, systems such as Astra Gemini, the demands on Target Fabrication to improve productivity and quality increase. It is essential to be able to trace, track and record target fabrication processes both during and after an experiment, to interrogate data and to understand the underlying science that is achieved. To deliver this service and to improve and streamline the processes from inception to delivery of a product the Target Fabrication Group has introduced a Quality Management System (QMS) into its experimental delivery procedures. This quality management process incorporates the full process of target fabrication from the initial conceptual design of a target, through the planning stage to manufacture and finally delivery to the experimental user group for their experimental shots.

What is a Quality Management System?

A QMS does not just focus on the quality of the product but also focuses on the means to achieve quality. It is made up of procedures, processes and resources which collectively monitor and improve the system. This reduces the risks involved whether financial, safety or consistency of work. A QMS provides the framework for continual improvement to increase the probability of enhancing customer satisfaction¹. The principal concepts of the QMS are²

1. Achieve, maintain and seek to improve continuously the quality of its products in relationship to the requirement for quality.
2. Improve the quality of its own operations, so as to meet continually all customers and other stakeholders stated and implied needs.
3. Provide confidence to its internal management and other employees that the requirements for quality are being fulfilled and maintained, and that quality improvement is taking place.
4. Provide confidence to the customers and other stakeholders that the requirements for quality are being, or will be, achieved in the delivered product.

5. Provide confidence that the quality system requirements are fulfilled.

Benefits

Many of the systems used in a QMS occur in the natural development of any department – any business that has run for more than 3 years will already be 80% compliant, and Target Fabrication was no different. There were very few changes in procedures that were required to fully integrate a QMS into Target Fabrication, but the benefits have had a very high impact, not only to Target Fabrication, but also to the communities that interact with them.

Traceability of Targets There is now complete traceability of the materials used in each target. When an external supplied component or material is ordered it is recorded in a central order book that all of Target Fabrication staff can access giving all incoming materials a unique number. The numbers are recorded against each target identification number so every part of each target can be traced back to its source. (The order book is also used for tracking non-target related items as part of the QMS procedures). This small addition to the purchasing procedure gives a huge benefit to both Target Fabrication (in terms of tracking batches of parts) and also to users who have confidence that their targets (and parts) meet good quality requirements. Any anomalous results can be traced back, for example, to a possible supplier issue. The tracking also gives assurance the correct targets have been manufactured and delivered. The recording of targets and their components also means that it is easier for Target Fabrication to keep track of completed orders if a change of fabricator is required part way through an experiment. It is now much easier to share or delegate the fabrication of targets if the fabricator is off-site or required elsewhere. All procedures are now recorded and approved and this allows for more consistent fabrication as all fabricators now work to the same defined methods. Significant effort has been focused to ensure that although the QMS is thorough it incorporates the experimental nature of the department and has been designed to cope with the daily changes that are inherent in the work that is carried out while still maintaining accurate records.

Changes made to the Target Fabrication Structure

As part of the new system, two new roles were identified - a Laboratory Quality Manager (QM), who oversees the system and makes sure that the procedures are helpful rather than a hindrance; and an internal auditor, someone to inspect how well the system is being followed. The QM monitors the results of all processes such as on-time-delivery, and the number of non-conforming (defective) targets produced. All changes are reviewed and, where possible, procedures are put in place to improve results. The QM also monitors results from the coating plants, customer feedback, and internal audits.

Internal Audits

Whilst the procedures that have been defined in the QMS are fixed, the department itself is constantly changing. The task of an internal audit is primarily to ensure that the procedures still meet the requirements of the department. Target Fabrication activity has been split into 12 different areas; each area is audited annually by the internal auditor. Changes that have been made already include the move to using Shared Services for purchasing, and the need for enhanced documentation during the planning phases of experiments.

External Audits

To be granted accreditation the Target Fabrication QMS has to be inspected by an external ISO9001:2008 certified auditor. This inspection is split into 2 parts. The documentation audit was carried out in March 2010 and there were found to be no Major problems. The final certification audit will be in August 2010.

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Electroplating of gold and palladium for high power laser target fabrication

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Introduction

This article describes the setting up of gold and palladium electroplating processes in the Target Fabrication Laboratory. This will meet the needs of the user community for: thick metal layers (up to 30µm), coating of complex intricate forms and different types of metal surface finish. In the past thick films were deposited using coating plants in a long and expensive process or subcontracted.

Electroplating process

Electroplating (also called electrodeposition) is the process of producing a coating on a prepared conductive surface by the action of an electric current. The deposition of a metallic coating onto an object is achieved by putting a negative charge on the object to be coated and immersing it into a solution which contains a salt of the metal to be deposited. In other words, the object to be plated is made the cathode of an electrolytic cell (Fig. 1). The metallic ions of the salt (M^+) carry a positive charge and are thus attracted to the object. These metal ions receive electrons and are reduced to metallic form at the interface between the solution and the cathode, plating onto its surface [1, 2].

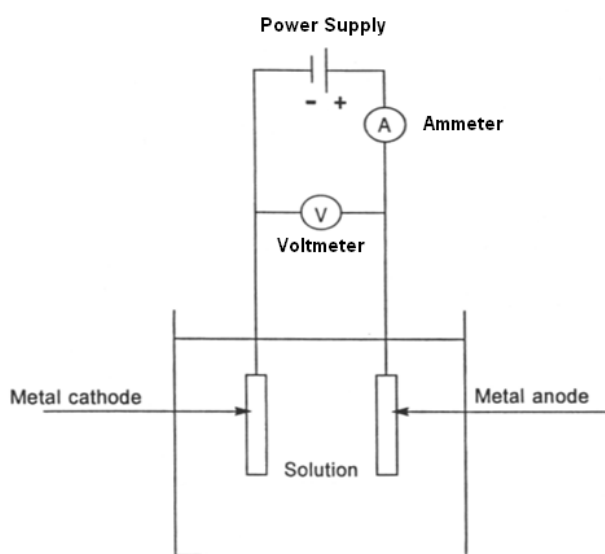


Fig 1: Schematic for electroplating.

Sophisticated plating electrolyte formulas have been developed and are being routinely employed providing greater control over the deposition process such as layer thickness, porosity, adhesion and surface finish. New developments enable greater plating speed and better throwing power (the ability of a plating solution to produce a relatively uniform distribution of metal on a cathode of irregular shape). In addition, electroplating of materials such as palladium, gold and copper are now broadly used in electronics for connectors, circuit boards, contacts, etc. Generally the more complex the ion the more efficient and smoother deposits occur compared to simple ions [3].

In aqueous solutions such complex ions dissociate slightly into simpler ions, and metal is deposited by direct discharge from the complex ion.

Compared with other metals, pure gold is chemically least reactive, though it is attacked by aqua regia (1:4 volumetric ratios of nitric acid and hydrochloric acid solution) and can be dissolved in alkaline solutions of cyanide or sulphite. Most gold electrolytes are of the complex cyanide ion that efficiently creates smooth deposits. However, to avoid using cyanide, which is highly toxic, an electrolyte based on ammonium gold sulphite was used. Compared to cyanide electrolytes, the sulphite process produces more bright, hard and ductile deposits. Another advantage is the good throwing power of these electrolytes. The disadvantages are the limited life due to formation of free sulphite, leading to a rapid increase in solution density and precipitation. Another drawback is the higher commercial cost of the sulphite solution due to its complex preparation [4].

Palladium is a greyish white metal that is attacked by nitric acid. It has high corrosion resistance though it oxidizes fairly easy when heated. It is unaffected by most acids, alkalis or other corrosive agents with the exception of aqua regia [5]. For our palladium plating a virtually neutral solution based on palladium diammino-nitride complex was employed.

Electroplating experiments

In order to set up and optimize the plating processes for Au and Pd initial experiments were conducted on copper foils. Flat samples were chosen to facilitate area calculation, sample preparation and surface characterization.



Fig. 2: Plating setup in Target Fabrication.

The influence of current density and surface cleanliness on the adhesion and morphology of the plating deposit was studied. This was done while maintaining constant agitation, solution pH, temperature, anode type and distance between electrodes. After being optimized, the gold plating process produced a smooth deposit with good brightness and ductility (fig. 3). The coatings thickness, morphology and roughness were characterized by SEM and surface profilometry. A good throwing power was observed: the surface roughness tended to decrease as the plated deposits thickness increased.

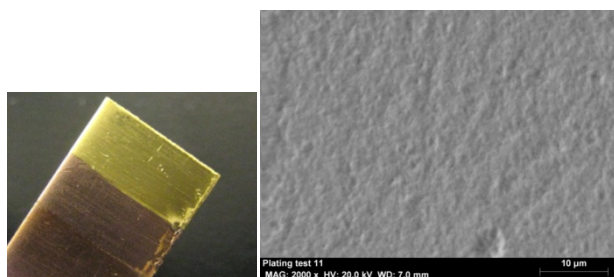


Fig. 3: Optical and SEM images of electroplated Au.

Electroplated palladium grew into more nodular coarse deposits and the throwing power was not as good as gold (fig. 4). However the roughness and adhesion of the deposit were good enough for our intended applications.

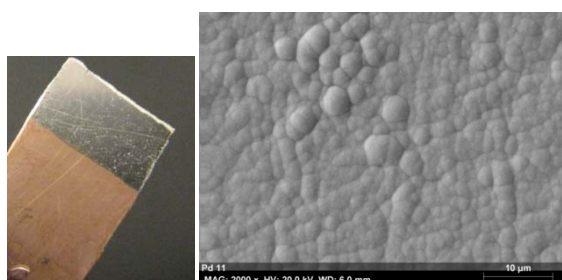
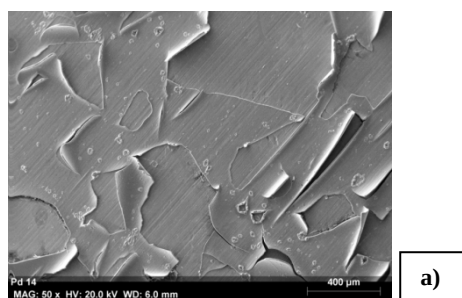


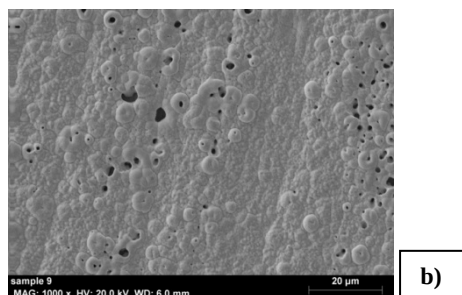
Fig. 4: Optical and SEM images of electroplated Pd.

The palladium and gold electrolytes require regular maintenance by adding ammonia solution when the pH drops and filtering when solid particles are found in suspension. The maximum thickness obtained for Au deposits was 35µm. The Pd processes allow us to obtain metal coatings that can go up to 25µm thickness. However it may be possible to go beyond these thickness limits by using very low deposition rates.

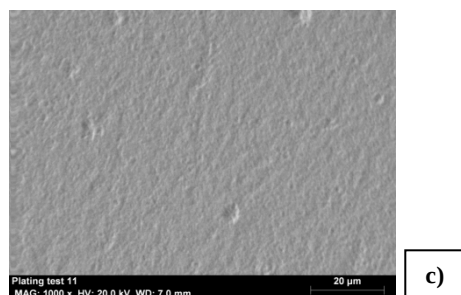
As expected, the surface preparation had a strong influence on the quality of the plated deposits for both Au and Pd. Substrates that were only cleaned with detergents and solvents under ultra sonic agitation and heating yielded deposits that peeled off in some areas, especially under nitrogen clean or by pull out tests with adhesive tape (fig. 5a). It seems that although organic compounds such as grease or oil were removed during cleaning there was still surface oxide that weakly bonded with the metal deposit.



a)



b)



c)

Fig. 5. SEM images of plated deposits: a) substrate cleaned with solvents and detergent, b) acid cleaned, c) acid cleaned and lapped.

By adding the step of cleaning the substrate with a diluted solution of sulphuric acid the deposit adhesion improved although some porosity, nodular growth and small areas of bad adhesion remained (Fig 5b). Finally, a third cleaning step of lapping was added where a more compact fine grained coating was plated. It is evident that a smooth lapped surface, free from scratches and pitting is more favorable for plating. It is also possible that the acid clean on its own was not totally effective at removing the oxide (Fig 5c).

Conclusions

Initial steps have been taken to develop a new deposition process at Target Fabrication. This process is quicker and more affordable for the deposition of thick metal films. However it is limited to conductive surfaces that are chemically resistant to the plating solutions. As a result, any incompatible surfaces should be coated with a thin conductive seed layer prior to plating.

Although only individual samples were plated this process can be adapted for mass production by making adjustments such as a larger plating vat or a suitable sample holder. For the deposition on non conductive complex forms, such as polycarbonate spheres, more work must be carried out. In the future, other metals such as Cu or Ag could be added to the Target Fabrication plating capability.

Acknowledgements

The authors would like to thank the support from John Spencer from the Space Science and Technology Department (SSTD) and also Gabriel Schauman from GSI Darmstadt. Also we acknowledge the contribution from the Laser for Science Facility (LSF) and the CLF Mechanical Workshop.

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Mass Production of AFI Cone Geometries for Fusion Target Studies

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Introduction

The recently launched HiPER (High Power laser Energy Research facility)¹ project is a European initiative to open up Inertial Fusion Energy as a commercial process for energy generation. One of the baseline targets² for the HiPER project uses an innovation called AFI (Advanced Fast Ignition) in which a fuel sample is compressed using multiple laser beams around the end of a gold cone. An 'ignition' beam is then injected into the cone to induce a thermo-nuclear reaction. The HiPER facility is planned to run at a repetition rate of up to 10Hz and will require a large number of targets. If the baseline target selected is an AFI target then there will need to be a reliable supply line of high quality high specification target components at a low cost (a few cents per target). This report summarises the current capabilities of cone production in STFC and gives consideration to the scale up options that are available.

Standard AFI Cone Production

The first demonstration of the AFI concept was at the Rutherford Appleton Laboratory in 2000³. The cone targets for this experiment were produced in a 'one-off' fashion where they were turned on a lathe to profile the inside and outside features⁴. This production process supplied high quality targets but in low numbers and the cost of an individual target was upwards of £1000. While this was a feasible production method when experimental requirements were low, to carry out statistical studies and investigate a number of different parameters upwards of 50 targets are needed for an experiment. Therefore to increase the number of targets that are available the development of techniques for the mass production of cone targets was investigated.

Simple-geometry cone targets were produced using a high precision Kern CNC micro-milling machine that is capable of reproducing the geometries available on a lathe. The fully automated machine with online tool wear measurement and tool change capabilities produced a number of mandrels ranging in number from single cone geometries to arrays of up to 25 targets. This technology was scaled to be able to produce 50 cone targets in a production run opening up the way to statistical studies during experimental campaigns⁵. (See figure 1)

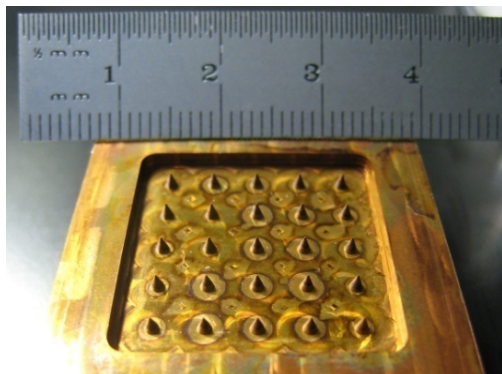


Figure 1: A 5 x 5 machined array of microcone mandrels

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The targets that were produced in this way were shot on the Vulcan Laser system in November 2008 and exhibited a good performance⁶. The surface roughness of the internal walls of the cone was less than 1µm Ra. (See figure 2.)

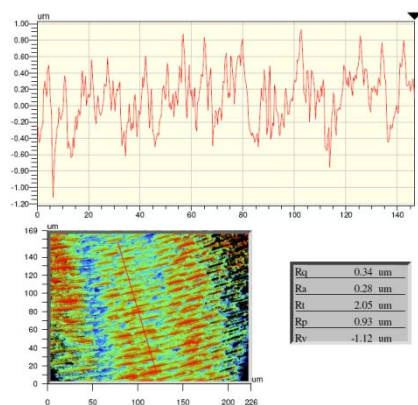


Figure 2: Lineout and 2D data showing the surface roughness on the internal wall of a mass-produced cone

Other Materials

Using the above production method it is also possible to produce cone targets in copper. By machining an aluminium mandrel and plating with copper it is possible to mass produce copper cones in batch numbers of 50. (See figure 3)

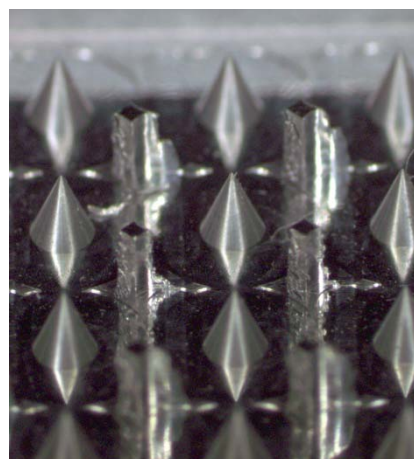


Figure 3: An array machined in aluminium for copper plating

Advanced Geometries

Using similar techniques to the production of standard cone targets it was possible to mass produce cones with novel geometries. A formula for a parabolic shape was used to create a number of machining points which were input into the Kern machine. (See figure 4)

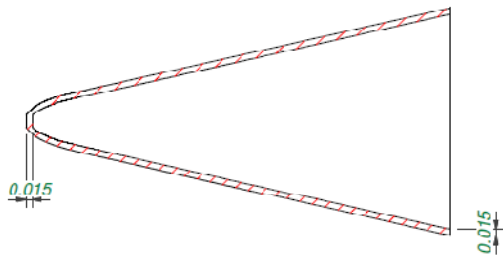


Figure 4: The design for the parabolic cone targets

Using a tool path that stepped every $2\mu\text{m}$ a mandrel was created in aluminium the replicated the internal profile of the cone. This was then plated in copper and re-machined using a number of datum points to ensure that the wall thicknesses were constant. For a number of cones the tip was removed to leave an open ended cone.

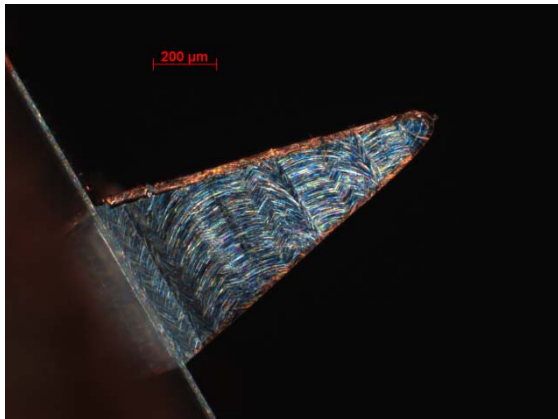


Figure 5: The sectioned copper cone on the sectioned aluminium mandrel

Figure 5 shows the reproduction of the parabolic shape on a cone that has been sectioned. To remove the aluminium mandrel from the rest of the cone the samples were placed in sodium hydroxide to etch the aluminium.

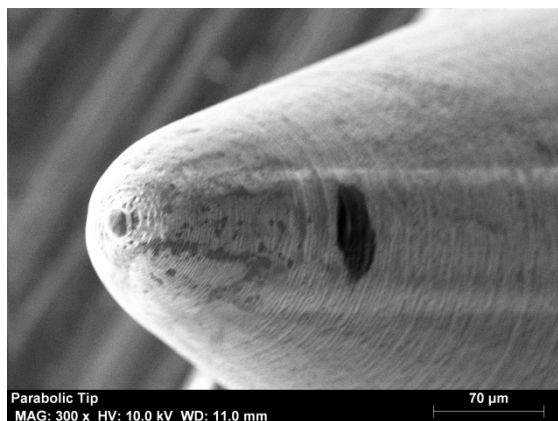


Figure 6: A SEM image of the parabolic shape of the tip.

Figure 6 is an SEM scan of the parabolic tip that shows the form and also gives an indication of surface roughness. The wall roughness is similar to that on a standard cone but as the step size is constant, the parabolic tip shows a smoothing effect since the tool interaction is different when machining the changing angle at the tip of the cone. There is also a small feature at the tip of the cone where the material has not been fully removed.

In figure 7 the tip of the cone has been removed while the mandrel is still in place. In this instance it can be seen that the hole size that will be left when the mandrel is removed will be in the order of 40-50μm diameter.

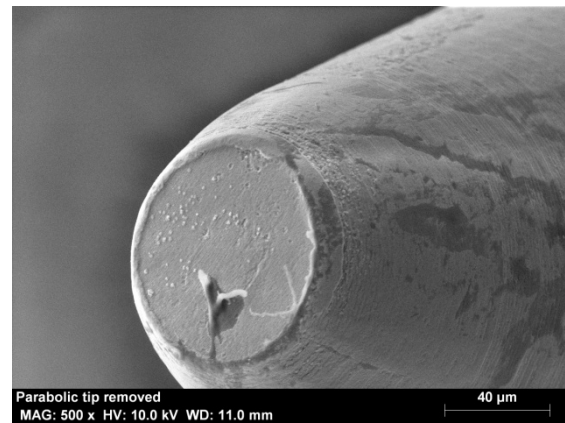


Figure 7: SEM image of a parabolic cone with the tip removed (mandrel still present).

Further Work

The next generation of cones will included a number of different material layers and locating features to aid assembly. Three layer cones will be made from chrome, aluminium and gold and are formed on a novel material for the mandrel. The targets are due to be fielded in late 2010.

Conclusions - Scale-Up to High Rep Rate Systems

While a significant step change has been introduced in moving to mass production in batches of 50 this cannot be scaled to the numbers required for a HiPER style reactor. The reported technology clearly opens the way to 100-1000's of shots but another step change in the manufacturing is needed to deliver to a reactor. Such technologies could include hot pressing or wafer based production but whichever technology is chosen will clearly need to be fully integrated with an assembly station and also cryogenic filling and layering stations and, eventually, a full injector system to give an integrated target solution.

Acknowledgements

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Debris Analysis and Mitigation for Target Motion Systems

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Introduction

Laser-target interactions can create a large amount of debris which has the capability to damage optical components within the interaction chambers. Historically, the main protection has been towards the optical components due to the high cost of replacement. The target debris also creates issues for other components within the interaction chambers, which may not be expensive, but are part of the critical operating systems for the facilities. This paper documents the recent investigations into the effect of target debris on the motion systems used in the HPL facilities.

Debris Analysis.

Almost all x,y,z motion systems in use within the experimental chambers on Vulcan and Astra facilities use DC motor systems and Sony Magnescale encoders for position referencing. The only exception to date is the new Gemini inserter system which uses a hexapod design. The standard mounts, commonly referred to as 602's after the original drawing number have been the 'workhorse' mount style for over 10 years, and the associated technology within the mounts has shown exceptional reliability and functionality. Over the last few years however, the number of problems associated with drive failures has increased. Several factors have contributed to this, ranging from EMP damage in motion controllers to cable faults due to poor cable management. One failure type which has been a re-occurring theme of late is the 'sticking' of the Sony Magnescale encoders, where the Cu-Ni-Fe alloy, magnetically encoded rod becomes jammed within the read head. Figure 1 shows the standard produce in use, which has a clearance between rod and head of only a few 10's of microns in order to achieve the 0.5µm measurement resolution.



Figure 1. Sony Magnescale rod as used on the existing 602 encoded motion systems.

This 'sticking' if the rod has been the most common failure mode over the last 2 years, and it is usually fixed by replacement of the magnetic rod in-situ. For complex experiments, this usually means attempting to access the mount whilst negotiating the surrounding diagnostics, and since the 602 design is both compact and low-profile, this replacement usually takes around 30 minutes for an experienced person.

Analysis of rods post-experiment has helped to determine the main cause of this 'sticking', which was at first believe to be a misalignment of the read head and rod itself. The

analysis has revealed instead that the main cause is from target debris coating the rod, and in places depositing large 'blobs' of target debris. Figure 2 shows one of the images of target debris on one of the rods which caused failure in one of the target mount motions.

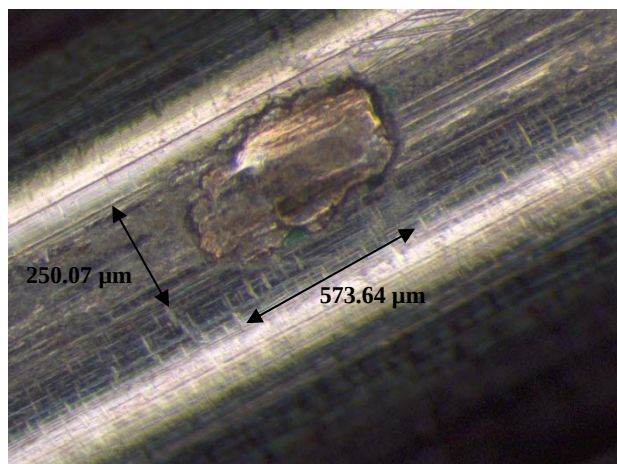


Figure 2. High-resolution optical microscope image of target debris on the Magnescale rod.

The debris observed on the mount is in the order of a few hundred microns in size, and is much larger than the clearance between the rod and read head on the encoder. Tests to determine the contact strength of the debris confirm that it is effectively welded onto the rod surface and is almost impossible to remove without inflicting damage to the encoded rod itself. An Energy Dispersive x-ray Spectroscopy (EDX) image of the section containing the debris allowed the identification of the elements present. This image was taken using the capabilities within the CLF's Target Fabrication Laboratory.

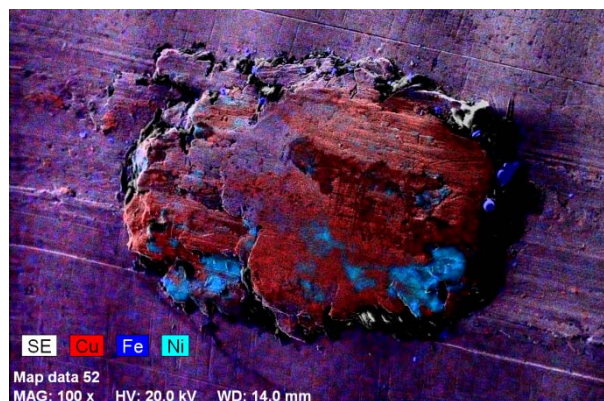


Figure 3. EDX image of target debris coated onto the magnetic rod.

The debris analysis shows a large lump of mainly copper debris welded onto the Magnescale rod. There are also traces of nickel

and other less obvious elements within the debris, all of which can be traced back to target materials used.

Debris Mitigation.

In order to cover the encoders to mitigate against debris, two routes are being investigated. The first is the replacement of all the encoders with enclosed versions of the same technology. This is not ideal, since the enclosed rod versions are much bulkier and would require a major redesign of the mounting system. The associated cost of replacing the encoders on over 20 mounts, each with 3 axis is also fairly significant. The second, preferred route is to add compressible covers onto the rods, which will allow the full freedom of motion without having a significant impact on the design of the mount. Although a final product decision is yet to be made, an example product range is shown in figure 4.



Figure 4. Flexible moulded bellows being investigated for covering the magnetic rod.

Conclusions

The majority of failures of target motion systems over the last few years have been observed to be linked directly to debris production from the laser-target interaction. This finding has enabled the development of the existing 602 3-axis target mounts to be directed towards the mitigation of debris and thus dramatically reducing the expected number of failures in the future. Protection method and hence production type will be directly controlled by the need to maintain a compact mount design as well as minimize the associated cost impact.

Acknowledgements

The authors would like to acknowledge the support of Axis (www.axis-gb.com) who have supplied the CLF with the Sony Magnescale encoders for many years and have been directly involved in the process of identifying the recent failure mechanisms.

Calibration of Image Plate response to energetic Carbon ions

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Introduction

A wide range of detectors has been developed to reveal the different existing types of radiation. Among those there are solid state detectors such as CR39, which is used as a nuclear track detector for particle counting; films with an active medium consisting of an emulsion (e.g., radiochromic films (RCF)); photodiodes; CCDs; photomultipliers (PMT); electron multipliers (EMT); microchannel plates (MCP); probes such as Faraday cups; scintillators (glasses, plastics, gases, liquids, crystals). Scintillators use the principle of photoluminescence (PL) that occurs when an atom is excited to a higher energy state and then returns to a lower one at a later time. PL is characterized by two forms of luminescence: fluorescence and phosphorescence. Typically a scintillator is made of fluorescent material and releases the energy in a short time (less than microseconds). The image plate (IP) is made of phosphors with phosphorescent properties which can release the stored energy in a long-lasting de-excitation (few hours). The IP is fabricated depositing BaFX:Eu^{2+} ($\text{X}=\text{Cl}, \text{Br}$) crystal grains on a polyester support film. The grains have a size of about $5\mu\text{m}$ and are kept together by means of a urethane resin [1]. The energy stored in the IP can be retrieved by stimulating the excited metastable state. The stimulation can be done by photons, and then the energy is released as light and called photo-stimulated luminescence (PSL). The IP is read using a scanner. The surface of the IP is scanned by a laser diode that de-excites the metastable state generating UV light which is read by a photomultiplier tube (PMT) and then converted in an electric signal. The output signal of the PMT is proportional to the light illuminating it. The signal is digitized by an analog-to-digital converter (ADC) and an algorithm in a 16bit discrete number. This quantization allows to store the signal as a 16bit TIFF image, in which each pixel represents a scanned position on the IP surface. The spatial resolution is typically $25\mu\text{m}$ or $50\mu\text{m}$, but some scanners can go up to $5\mu\text{m}$ resolution. The IP has a high sensitivity to the stimulating radiation and the response is linear [2]. The IP dynamic range is very high, but the scanner settings can give a reading of five orders of magnitude at best. In any case, the IP can be scanned more than once if necessary. Once used, the IP can be erased by exposing it to intense light for at least 15min.

The response of the Image Plate detector to protons has been reported in earlier works [3,4]. We discuss here some measurements in which the IP response to Carbon ions has been measured by cross-calibration with CR39.

Experimental set-up and methods

The measurements were carried out in the Target Area Petawatt (TAP). The VULCAN laser operates in the infrared range at a wavelength of 1053 nm . The laser pulse is compressed using the Chirped pulse amplification (CPA) technique obtaining pulse duration of sub-picosecond ($\sim 700\text{ fs}$). The laser beam was focused onto a thin metal foil, typically Cu or Au by using an $f/3$ off-axis parabolic mirror and a plasma mirror. The laser pulse energy typically used is about 450 J in a $\sim 10\text{--}20\text{ }\mu\text{m}$ focal spot. The laser energy losses due to compressor gratings and

plasma mirror were measured using a calorimeter, and they are 50% and 20%, respectively; giving a total energy loss of 40%. and a laser intensity on target up to $3\cdot 10^{20}\text{ W/cm}^2$. Such an intense laser pulse can generate proton and ion beams by means of the Target Normal Sheath Acceleration (TNSA) mechanism [5]. These ions originate from hydrocarbons and water vapour impurities usually present on the target surface. Owing to this acceleration mechanism broadband particle sources are generated with energy cut-off up to tens of MeV/nucleon [6]. A Thomson Parabola (TP) was used as mass spectrometer to discern the different ion species present in the ion source. The TP was mounted in axis with the laser and placed at the rear side of the target. The distance between target and TP pinhole was 630 mm . IP and CR39 were used as detectors (see Fig. 1). In order to calibrate the dispersion power of the spectrometer, the IPs were filtered to obtain a known energy cut-off of proton by performing a few laser shots.

A comparison of the cut-off positions along the dispersion plane with the analytical calculations gives a relationship between ion energy and position on the detector.

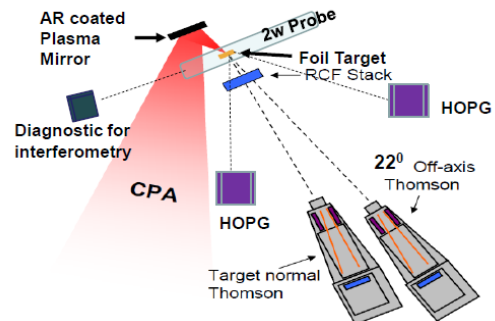


Figure 1. Layout of the experimental set up

IP characteristics

IP from Fuji Photo Film Co. Ltd is composed of a photostimulable phosphor layer made of very small crystals with a grain size of about $5\mu\text{m}$ and a density of 5.2 g/cm^3 . Among several types of IP the BAS-TR2025 has been chosen in this experiment. For this type of IP the phosphor layer thickness and density are $50\mu\text{m}$ and 2.85 g/cm^3 , respectively [7]. Its main feature is the absence of the plastic protective layer usually present in the most of the other types of IPs. The purpose of the layer is to preserve the response efficiency of the phosphor active medium by avoiding its degradation. However, it also slows down the fast ions and can even stop low energy ions before they reach the active phosphor layer, then making them undetectable. The typical thickness of the protective layer is about $10\text{ }\mu\text{m}$. This thickness is sufficient to stop protons and carbon ions with energy per nucleon below about 800 keV/amu . The IP can store the energy received by the impacting particles owing to the excitation of a metastable state of the phosphor, but on the other hand and for the same reason it has an intrinsic fading out time. Experimentally, it has been shown that the

phosphor releases the energy with two different decay times [2,3].

The metastable state de-excites first with a fast decay time losing about 20% of the stored signal in 20min and then with a longer decay time bringing the initial signal to 60% of its value in about 3hr.

CR39 characteristics

The CR39 is a plastic polymer used as solid state detector in nuclear science to detect ion, proton and neutron; it is insensitive to other forms of radiation. These particles leave trails when they travel through the bulk material and deposit their energy slowing down according to the stopping power theory. A typical CR39 plate has a thickness of about 1 mm and density of 1.3 g/cm^3 . 1mm thick CR39 is able to stop (and detect) proton and Carbon ion with energy up to 10 MeV and 230 MeV, respectively. The tracks left in the bulk are revealed by etching the CR39 in a sodium hydroxide solution. As the etching time increases the tracks get larger, creating pits on the surface. The etching was made in a 6.0N NaOH solution at a temperature of about 70°C for 15 minutes. Throughout the etching and counting procedure attention was paid to get pits clearly visible and avoid overlapping.

Result

In order to obtain the calibration of the IP BAS-TR2025 for Carbon ions of different energies a TP is used to disperse the ion beam onto the CR39 and IP. A slotted CR39 placed on the IP was used as detector so that carbon ions having reasonably close energy can be recorded at the same time around the edges of the slots, both on the CR39 and IP. After etching, the CR39 was observed with an optical microscope. Images of the pits distributed along the dispersion axis were grabbed using a CCD and then the pits around the edges of the slots were easily counted. Below, in Figure 2, an image representative of the pits' trace in the region of the slot edge is shown.

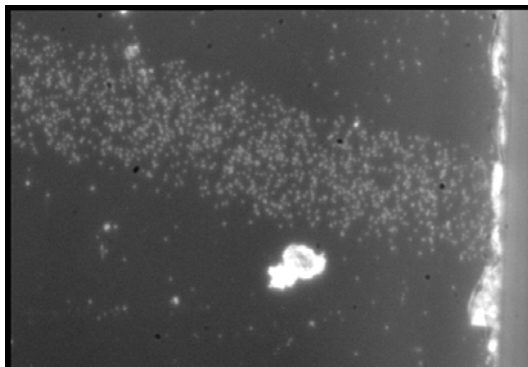


Figure 2. Image of the pits produced by Carbon 6+ ions in the region of a slot edge of the CR39 (right-hand side). The image is taken with a magnification 10x. The analysis is done using greater magnification.

The IP was read using the Fuji Film FLA-5000 scanner with a scanning pixel resolution of $50\mu\text{m} \times 50\mu\text{m}$, sensitivity of 5000 and latitude of 5. The output is a TIFF image that holds information about the PSL strength in a 16bit integer number. Using the algorithm provided with the scanner specifications it is possible to get back the PSL strength per pixel, which is proportional to the energy released by the particles hitting a particular pixel area. The relationship between particle energy and position on the detector is obtained by using a code in which the motion of a particle passing through the TP is calculated. Figure 3 shows the IP scan image used for the calibration. A Cu target 500 nm thick was used in this shot.

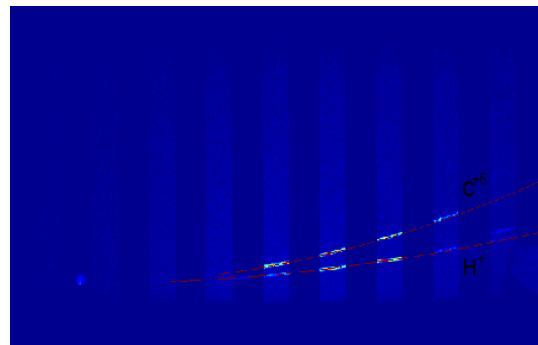


Figure 3. IP scan image showing the shadow of the CR39 slots and proton and carbon +6 ion signals. The red curves are the theoretical fits obtained using the above-mentioned code.

Owing to the code, dN/dE(E) , i.e the number of pits per unit energy and PSL/dE(E) , i.e. the equivalent PSL strength per unit energy, are obtained as a function of the carbon energy E. Each experimental point represents an edge of the CR39. In this way, it was possible to obtain 9 data points for fully ionized carbon ions. The calibration curve is the ratio of PSL/dE(E) to dN/dE(E) that gives the PSL value per carbon atom as a function of the carbon energy, $\text{CAL} = \text{PSL}/\#C(E)$. The function CAL(E) was multiplied by a correction factor to take into account the fade out time due to the spontaneous de-excitation of the IP phosphor. The correction factor is obtained from Ref.2, for the same type of IP.

Figure 4 shows the calibration curve for carbon ion with energy up to 40 MeV. The peak of the sensitivity curve appears to be around 27 MeV.

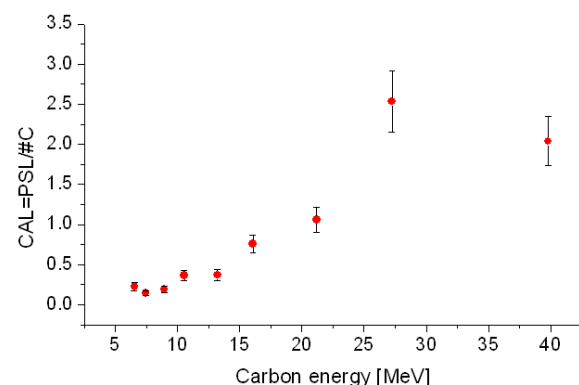


Figure 4. Calibration curve that relates the PSL signal to the number of carbon ions as function of the ion energy.

Conclusion

The absolute sensitivity of the IP BAS-TR2025 has been calculated by comparing the PSL value read by the scanner Fuji Film FLA-5000 to the number of carbon ions estimated by using the CR39. The readout value can be slightly different depending on the readout system [3,4].

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