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**Low-Temperature
Calorimetry for Ionising
Radiation Dosimetry**

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Hugo Palmans,
Thorsten Sander and Simon Duane**

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Low-Temperature Calorimetry for Ionising Radiation Dosimetry

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ABSTRACT

The report considers in some detail the likely future application to Radiation Dosimetry of the development of various microwave resonance based calorimeter designs. The predicted sensitivity of microwave resonance calorimetry makes it clear that such a system can be competitive with conventional calorimetry, even at room temperature. Much greater advantages accrue if cryogenic operation can be implemented in an ionising radiation application. In addition a number of other possible longer term applications of these technologies have also been outlined which should be considered for future collaborative work between DEM and DQL scientists.

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CONTENTS

1. Executive Summary:	1
2. Introduction	2
3. Possible Cryogenic Calorimetry for Radiation Dosimetry	2
3.1 <i>General</i>	2
3.2 <i>Existing applications of low-temperature calorimetry</i>	4
3.3 <i>Novel microwave methods for low-temperature calorimeters</i>	4
3.3.1 Basics of Microwave Resonator Methods	4
3.3.2 Microwave Resonance Thermometry	7
3.3.3 Whispering Gallery Microwave Resonator	8
3.4 <i>Potential new applications</i>	9
3.5 <i>Novel temperature sensors for cryogenic calorimeters</i>	10
3.6 <i>Non-contacting High Sensitivity Thermometry</i>	10
3.6.1 Simple Microwave Resonance Thermometer	11
3.6.2 Whispering Gallery Microwave Resonator (with absorber insert)	11
3.6.3 Coupled Dielectric Resonators	12
3.6.4 High Q Mechanical Resonator Method	15
3.6.5 Magnetic Susceptibility Thermometer	16
3.7 <i>Ionising Radiation Considerations</i>	16
4. Summary of Materials Properties	17
5. Expected Performance from Microwave Calorimeter	20
6. Future benefits of the work to NPL	21
• <i>Innovative application of cryogenic technology to dosimetry standards</i>	21
• <i>Development of more sensitive calorimeters</i>	21
• <i>Development of a microdosimetric calorimeter</i>	21
7. Conclusions	22
8. Bibliography	23

1. Executive Summary:

Following several lab visits and discussions over the past 24 months between DEM (Quantum Detection Group (QDG)) and scientists from DQL (Radiation Dosimetry team (RD) previously known as CAIR) it has become clear that RD has a short term need for accurate calorimetry which we believe dielectric microwave resonator metrology, developed in QDG, can address. As a result of these discussions a deliverable (3.2.3) was included in the QM2004-7 programme for project 3.2 which called for a report on the applicability of single particle spectroscopy to ionising radiation metrology, with particular emphasis on how further developments of these devices could contribute to the NMS programme in that area. This report is the product output of the deliverable. The specific milestones incorporated in the QMP document are as follows:

- ❑ Identify the requirement of CAIR (now RD) for single particle detection and spectroscopy in ionising radiation metrology
- ❑ Describe further developments of these devices which will address the NMS programme requirement in that area

In addition we have drafted a SR proposal with RD scientists to pursue this work. The RD future single particle detection requirements will be assessed at a future meeting which we will call soon. In addition to single particle detection their main concern is with stopping power (i.e. the ionising radiation term which approximates to quantum efficiency) of any detector we design for them. We aim to address this experimentally in a future programme, either Quantum Metrology or Ionising Radiation.

2. Introduction

Discussions between members of DEM (Quantum Detection Group) and DQL (Radiation Dosimetry team) scientists identified two areas of immediate relevance to their requirements.

- To push calorimetry to lower dose levels
- Calorimetry is the preferred method because it measures absorbed dose directly

Two separate quantities are of interest:

- -Absorbed dose which is a macroscopic quantity
- Linking dosimetry with biological effects requires an understanding of *micro-dosimetry* i.e. the energy deposited in a single event.

The experience of the QDG, through recent QMP projects, in the areas of bolometry/calorimetry and microwave resonance are highly relevant to these requirements since QDG has proposed combining microwave resonance with calorimetry to provide highly sensitive, reproducible total energy absorption calorimetry. The following sections outline the current and anticipated future needs of RD scientists relating to these issues while also outlining the science behind a number of proposed calorimetric methods.

3. Possible Cryogenic Calorimetry for Radiation Dosimetry

Following several lab visits and discussions over the past 24 months between QDG and RD scientists it has become clear that RD has a short term need for accurate calorimetry which we believe dielectric microwave resonator metrology, developed in QDG, can address. A joint SR proposal to pursue this work has been drafted but has not been funded. RD's future single particle detection requirements will be assessed at future meetings and this issue is not addressed in this report. Beyond improved calorimetry RD's main future concern is with radiological properties of any detector we design. We aimed to address this experimentally in the next Quantum Programme, as set out in the roadmap, but unfortunately this proposal will not be funded.

3.1 General

Absorbed dose to water is the quantity on which a radiotherapy treatment prescription is based and there is a consensus that absorbed dose standards are preferred for calibrating any radiotherapeutic beam or source [1]. The most direct method for measuring absorbed dose is calorimetry, which relies on the measurement of the temperature increase in a medium that absorbs energy from ionising radiation.

The temperature rises measured with calorimetry in radiotherapeutical beams are of the order 1 mK at room temperature and combined standard uncertainties well below 1% are required. Consequently, much effort has been concentrated in trying to increase the sensitivity of the calorimetry technique, aiming for a temperature rise sensitivity of

1 μ K. However, despite considerable efforts during the last decades, the sensitivity has increased by no more than a factor of two.

The energy spent per ionisation ranges from 30 eV to 1 keV for air whereas in a semiconductor such as Ge it is lower, in the region of 3 eV. There is an existing identified but unaddressed need to be able to make measurements at a range of length scales from the single cell through the nucleus down to individual DNA strands, since biological effects of ionising radiation are determined by the amount of energy transferred in single events on the cellular or sub-cellular level. The main source of information on such event distributions are now obtained from low-pressure gas-filled proportional counters [2]. The ability to perform such measurements in a microscopic or nanoscopic sample in the condensed state would be a very substantial improvement.

In present radiotherapy installations a typical large x-ray beam has a 10 cm x 10 cm cross section and a large penetration depth, whereas charged particle beams have limited range. For example a 60 MeV proton beam for ocular therapy has a range of 3 cm. For a 3 g graphite absorber a thermistor with 0.1% of the total mass is generally used as a temperature sensor. Low Z materials are preferred since they represent a closer match to human tissue.

The temperature increase for a given amount of deposited energy is inversely proportional to the specific heat capacity of the absorbing medium. Since at low temperatures the specific heat capacity of an absorber drops drastically (as T^3 for most electrically insulating materials), low-temperature calorimetry offers a potential method to overcome the sensitivity problem. For example, compared to the value at room temperature, the specific heat capacity of graphite is reduced by a factor of 10 at the boiling point of liquid nitrogen and by a factor of 5000 at the boiling point of liquid helium. Another generic advantage over conventional room temperature calorimeters is that more sensitive thermometers can be realised at cryogenic temperatures based on a variety of techniques, including microwave resonance, superconducting transition and magnetic susceptibility. Thermistors, which are traditionally used in calorimetry, also exhibit an increased sensitivity with decreasing temperature, by a factor of 3 at the boiling point of liquid nitrogen and a factor of 100 at the boiling point of liquid helium, although in the latter temperature range specialised cryogenic thermistors need to be used [3] and special techniques to measure the high (and highly temperature dependent) resistance of thermistors at low-temperature may be needed [4]. Generic disadvantages of low-temperature measurements are the added complication and cost of provision of the cryogenic environment and a requirement to isolate the calorimeter from room temperature by some means (generally a vacuum jacket) which will introduce some uncertainty in absorption arising from this isolating medium. Another point of concern is the equivalence of the radiological properties at these operating temperatures compared to room temperature.

One outcome of this report is a proposed project which will allow the evaluation of low-temperature calorimetry for dosimetry using three temperature sensing techniques (the microwave resonance and non-contacting thermometry methods are introduced below). The main aim of this project would be to obtain a proof of principle of dosimetry capability, which we cannot deliver at present and which will enable bids for funding of more specific applications.

3.2 Existing applications of low-temperature calorimetry

Low-temperature calorimetry is by itself not a new technique and is used for particle counting in radioactivity [5-7] and in high-energy physics [3] to determine the energy of single particles. But the quantity *absorbed dose* is a macroscopic quantity and involves a different technological approach. Previous attempts at establishing low-temperature calorimeters for absorbed dose resulted in strongly inhomogeneous and non-water-equivalent (mostly metallic) media. Schleiger et al. [8] describe the development of a prototype microcalorimeter for the measurement of absorbed dose. This microcalorimeter was operated at 77 K, a thermistor was used as temperature sensor and an aluminium absorber was used. Dose rates as low as 0.03 Gy/min were measured in a ^{60}Co beam. Noteworthy is also the development of a water ice calorimeter for photon dosimetry [9] which was tested in a therapy level ^{60}Co beam. Measurements gave a signal to noise ratio of about 300 for a dose of 5 Gy and the sensitivity of the ice calorimeter operated at -10°C was increased by a factor of two compared to a water calorimeter operated at 4°C . However, the disadvantage in using ice was that the diffusivity of ice is approximately ten times that of water at 4°C , making the measurement of absorbed dose to water at a point more difficult. No attempt was made to isolate a sample of ice from the surrounding to overcome this problem.

One of the challenges for this study is to propose techniques to perform low-temperature calorimetry in a water equivalent and/or homogeneous environment.

3.3 Novel microwave methods for low-temperature calorimeters

We propose here a number of possible non-contacting thermometer methods which are based on the sensitive techniques of microwave resonance and have potentially μK sensitivity at cryogenic temperatures. In order to compare different techniques it is useful to define a figure of merit, for the purpose of comparing the suitability of the proposed microwave resonance calorimeters for the variety of ionising radiation *fluence* measurements. It can be shown (see section 4) that the minimum detectable temperature rise measurable in unit bandwidth ΔT is given by

$$\Delta T = \frac{NE\tau}{CA\lambda\rho}$$

where N is the number of particles/s impinging perpendicular to a cross sectional area A , E being the incident energy of the particles, τ the response time, λ the absorption range, C the heat capacity of the absorber and ρ the density of the *absorber*. Although rather sophisticated coupled resonant systems have been investigated at NPL for nK temperature discrimination in this report we begin by considering only two, relatively straightforward, potential techniques.

3.3.1 Basics of Microwave Resonator Methods

A microwave resonator may be constructed in a wide variety of ways. The essential feature is that within some spatial region microwave radiation may be introduced and confined. The region may be a volume enclosed by a metallic surface, with suitable

coupling structure to introduce and or extract microwave power. Equally it may be a thin film planar structure with metallic thin films separated by one or more dielectric layers. A third type of resonator is less well known and consists simply of a dielectric structure (generally 3D in nature) with a microwave permittivity which is much greater than unity. The response of a microwave resonator to reflected or transmitted microwave power, as the microwave frequency is varied, is generic and characterised by a number of resonant frequencies $\{f_i\}$ at which there is a peak in the absorbed power. The peaks have typically a Lorentzian lineshape in the frequency domain, characterised by a second parameter, the frequency width $\{\delta f_i\}$ of the i th resonance. The quality factor of the i th resonance $\{Q_i\}$ is defined by

$$Q_i = \frac{f_i}{\delta f_i}$$

where δf_i is defined as the full width of the Lorentzian measured at the half power points.

The frequency of the resonant modes of the resonator types above all depend on the dimensions of the resonator. In addition the planar thin film types and 3D dielectric resonators have frequencies which scale inversely with dielectric permittivity $\epsilon(T)$ of the substrate or puck structure, for a given size. An order of magnitude estimate for the lowest frequency mode of a resonator with linear size L with resonant volume filled with material with effective permittivity ϵ (i.e. averaged over the resonator volume) is given by:

$$f_0 \sim \frac{c}{2L(T)\epsilon(T)^{1/2}}$$

where c is the velocity of light in vacuum.

The reason why microwave resonance is a much used measurement technique in a variety of metrological fields is that frequency is one of the most accurately measured physical quantities. This is augmented by the fact that typical Q values, even at room temperature, can be as high as 10^4 , rising to greater than 10^6 at cryogenic temperatures. This allows very small changes in resonant frequency to be accurately detected.

Microwave resonance thermometry could be done using any of the basic resonator types mentioned in the preceding paragraphs. Thus if the temperature of a metallic housing of a cavity resonator changes the material will change its dimensions due to thermal expansion and thus its resonant frequencies will change. Although the linear thermal expansion coefficients of metal at room temperature are typically in the range $1\text{--}10 \times 10^{-6}/\text{K}$ it is still possible to detect sub-mK changes since the resonant frequency of a cavity resonator (with typical room temperature Q value of 10^4) may be easily measured to ~ 1 part in 10^{10} . For thin film resonators and 3D dielectric resonators thermal expansion of the component parts will also bring about a change in resonant frequency but in addition the effective permittivity is likely to have a rather stronger

temperature dependence than the linear expansion coefficient, enhancing the effective frequency change with temperature df/dT in general.

We have already demonstrated, in a long distant project within the predecessor of the present Quantum Programme, how a metallic cavity resonator can be used as a thermometer. In fact in these measurements the microwave resonance frequency variation with temperature was used as a non-contact method to measure the coefficients of linear thermal expansion of phosphor bronze and niobium [10]. Fig. 1 shows a schematic diagram of the measurement system used in this system which is generic to most of the other microwave resonance systems discussed below.

As an illustration of the thin film resonator technique [11] we show a schematic diagram of a thin film microstrip resonator in figure 2a. This has superconducting electrodes (YBaCuO₇₋₈) with a zirconia (ZrO₂) dielectric. The same system as shown in fig. 1 was used to measure the resonator resonant frequency as a function of temperature allowing the effective permittivity change with temperature to be deduced, as shown in fig. 2b.

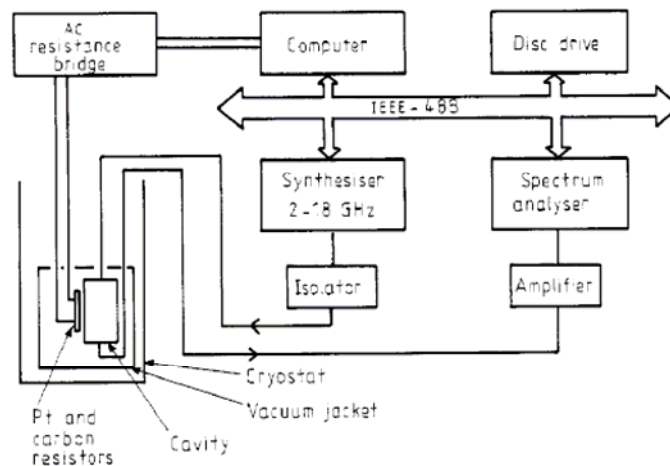
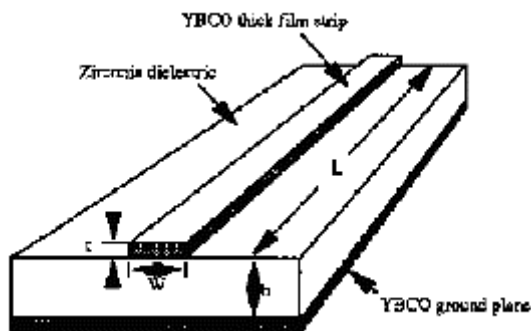
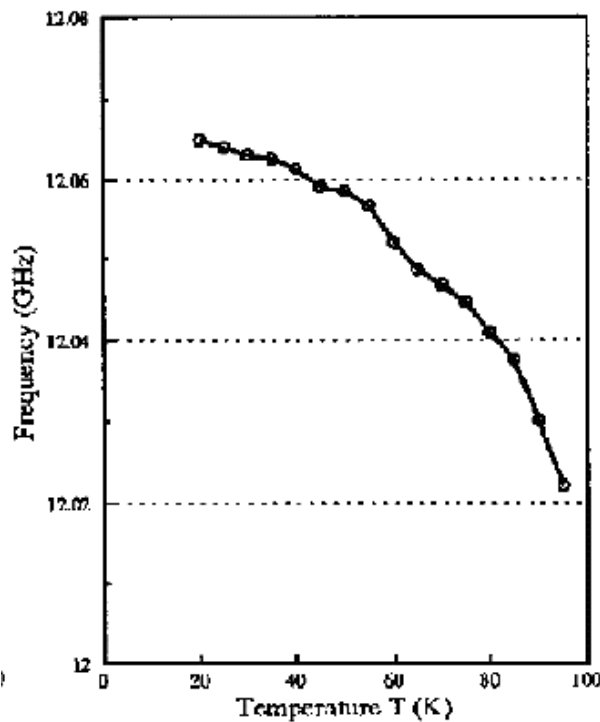


Fig. 1 Schematic diagram of microwave cavity resonance measurement system (see [10]).



(a)



(b)

Fig. 2 a) schematic of thin film microstrip resonator b) variation of effective permittivity of the microstrip resonator spacer with temperature, as measured by microwave resonance.

A third type of resonator uses a 3D sample of (usually single crystal) dielectric as the resonator. Provided the permittivity is much greater than 1 rather high Q values can be achieved without coating this dielectric 'puck' with a metal film to contain the microwave standing wave distribution. The mismatch in permittivities between vacuum (or air) and the dielectric of the puck is sufficiently large to allow high field confinement without the inevitable losses arising from induced microwave currents in any normal metal coating. Such a resonator (specially compensated to drastically reduce the effects of permittivity variation with temperature) is described in [12]. Fig. 3 shows a schematic of this type of structure.

3.3.2 Microwave Resonance Thermometry

The third method outlined above seems the most straightforward to satisfy RD's requirements. This treats the entire absorber as being made of a low Z, crystalline insulator with low microwave loss and reasonably strong temperature dependent permittivity. Then the resonator will form the reference element for a microwave loop oscillator whose oscillation frequency may be counted with high accuracy (frequency being the most accurately measurable physical quantity). Then when the absorber/resonator is exposed to a fluence of energetic particles these will be absorbed (totally or partially) within the volume, bringing about a temperature rise. This in turn produces a change in permittivity and thus a frequency shift of the oscillator which will be measured by the frequency counter. We believe that, even at modest cryogenic

temperatures such as 50K, ΔT may be made as low as 1 μ K using, for example, a rutile (TiO₂) resonator.

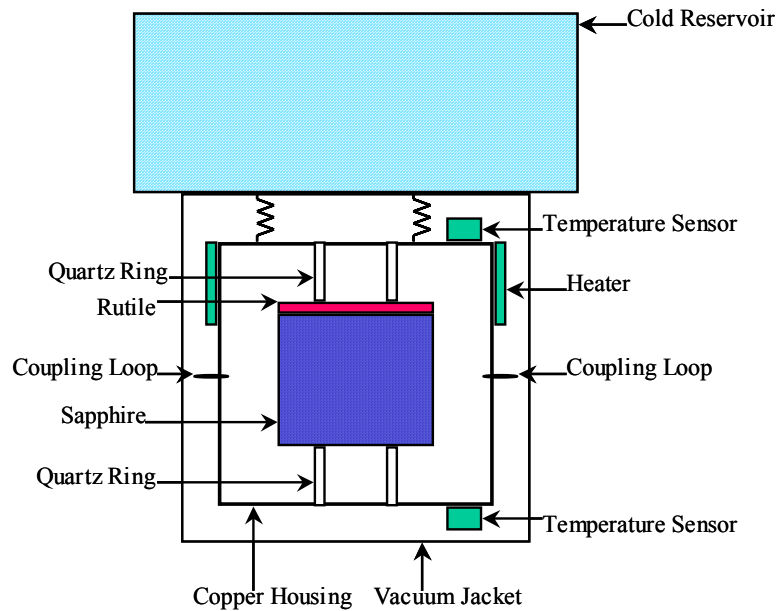


Fig. 3 Schematic of dielectric resonator (temperature compensated for thermal stability), see [12].

3.3.3 Whispering Gallery Microwave Resonator

The method of section 3.3.2 relies on the availability of a low Z material which satisfies the above conditions on permittivity, thermal properties etc. In the event that no suitable material is available an alternative, but related method, may be explored. This uses a somewhat counter-intuitive concept of separating the standing wave microwave field region from the absorber material. For high order resonant modes in e.g. cylindrical resonators there exists a class of modes (whispering gallery modes) in which the standing wave energy is concentrated around the perimeter of the cylinder and there is almost no energy stored on the axis (see fig. 4) [12]. This means that the quality factor of these modes is not much reduced even if the central region of the resonator is removed and replaced by a rather more lossy material. This central region could then be the ionising radiation absorbing element (e.g. graphite).

An additional advantage of all of these resonance methods is that there is no additional mass or electrical leads associated with thermometers. If one is prepared to go to the complexity of multimode excitation it is even possible in principle to read out the spatial variation of the absorber temperature allowing, in principle, some measure of the spatial profile of the radiation.

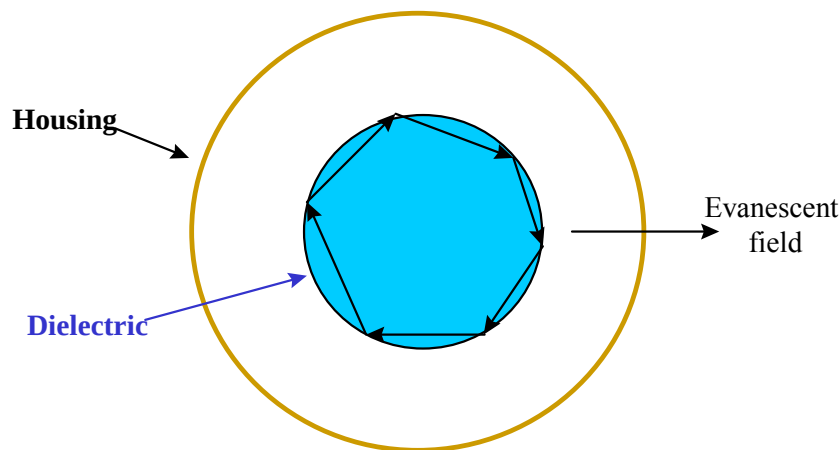


Fig. 4. A whispering gallery mode has a large number of nodes around the circumference of the cylindrical dielectric resonator. This leads to high ‘confinement’ within the dielectric (analogous to total internal reflection in optics).

3.4 Potential new applications

In the search for optimal treatment methods adapted to specific issues in diverse types of tumours and tumour sites, a variety of radiotherapeutical methods have been developed. Among these are a number of recently developed techniques that use dose rates which are one or more orders of magnitude lower than in conventional external radiotherapy treatment, such as low dose rate brachytherapy sources, radioactive seed implants and eye plaques. For conventional external treatments with high-energy photon and electron beams absorbed dose standards based on calorimetry have been established for a long time and have culminated in a number of international Codes of Practice for clinical dosimetry based on these standards. For internal sources, however, the only primary standards available at present measure *air kerma* at 1 m from the source with a standard uncertainty of 1%. The conversion of air kerma at this reference distance to absorbed dose at a point near the source results in standard uncertainties of 5 to 10% and in some cases even larger than 20%, which is too large a contribution to the uncertainty of the dose delivery in the patient. The major reason for the lack of absorbed dose standards for those types of sources is the low sensitivity of room-temperature calorimetry (compared to the measurement of ionisation in air). Despite this, the action plan formulated at a recent International Symposium on Standards and Codes of Practice in Medical Radiation Dosimetry organised by the IAEA, identified the need for research and development of calorimeters for brachytherapy. At present a room temperature calorimeter for high-dose rate brachytherapy is being built [13]. Also low and medium energy x-rays exhibit in general lower dose rates than required in room-temperature calorimetry and would gain from an increased sensitivity of absorbed dose calorimetry.

A second potential application to ionising radiation is related to the issue that not only the dose, but the relative biological efficiency (RBE) of radiation, is of importance in the outcome of a treatment. For high-energy photons and electrons RBE is constant, but

for photons and electrons with energies below 50 keV which are for example used for eye treatments, it could be different and for low-energy light-ions RBE definitely is different and energy dependent. The physical quantities that are related to these changing RBEs are microscopic energy depositions or so called microdosimetric quantities, which quantify the energy deposited per ionising event rather than per incident particle. Present day microdosimetry is performed with proportional counters that measure ionisation in a tissue equivalent gas cavity scaled by adapting the gas pressure to the size of a biological target, such as a DNA strand, a cell nucleus or a cell. Low-temperature calorimeters with very thin absorbers would allow an alternative to measure the energy deposited in such a single event on the real geometrical scale in water or a tissue equivalent material.

Apart from the above mentioned proof of principle, a feasibility study would provide a sensible future route to investigate the application of low-temperature calorimetry to dosimetry of brachytherapy sources, medium-energy x-rays, small fields (radiation beam sizes less than 4 cm x 4 cm) and to microdosimetry.

3.5 Novel temperature sensors for cryogenic calorimeters

In this and following sections we describe some of the non-contacting thermometry methods which we have imagined could be useful to ionising radiation measurements. Cryogenic calorimeters have two clear and generic advantages over conventional room temperature calorimeters:

- The heat capacity of an absorber of given size falls rapidly at low temperatures (as T^3 for most electrically insulating materials) so that the temperature rise produced by the absorption of a given energy is greater.
- More sensitive thermometers can be realised at cryogenic temperatures, based on a variety of techniques including, amongst others, microwave resonance, superconducting transition or magnetic susceptibility.

On the other hand they also exhibit some generic disadvantages:

- The added complication and cost of provision of the cryogenic environment
- A requirement to isolate the calorimeter from room temperature by some means (generally a vacuum jacket) which will introduce some perturbation arising from this isolating medium.
- Radiological properties for the materials in the calorimeter, especially the stopping power of the absorber [14] might be temperature dependent for which corrections need to be determined.

3.6 Non-contacting High Sensitivity Thermometry

There are several possible non-contacting thermometer methods which have potentially μK sensitivity at cryogenic temperatures. Combining this level of sensitivity with the reduced heat capacity of the absorber/resonator at these reduced temperatures gives the required improved sensitivity for calorimetry. A figure of merit may be estimated, for the purpose of comparing the suitability of the proposed calorimeters for the variety of ionising radiation fluence measurements: consider a fluence of N particles/s impinging perpendicular to a cross sectional area A with energy E per particle and absorption range λ the energy absorbed per second is NE in a volume λA . If the heat capacity per

unit mass of the absorber at the operating temperature T is $C(T)$ and the density of the absorber is ρ and the thermal link to a heat bath at T_0 is G then the steady-state temperature rise of the absorber will be ΔT where

$$\Delta T = \frac{NE}{G}$$

whereas the thermal time constant (i.e. the time it takes to approach the steady-state) is τ where:

$$\tau = \frac{C(T)A\lambda\rho}{G}$$

If our final figure of merit is δD , defined as the minimum detectable absorbed dose, measured in unit bandwidth, it may be expressed in terms of the above parameters and the minimum detectable temperature rise measurable in unit bandwidth δT :

$$\delta D = C(T)A\lambda\rho\delta T$$

3.6.1 Simple Microwave Resonance Thermometer

The first method treats the entire absorber as being made of a low Z , crystalline insulator with low microwave loss and reasonably strong temperature dependent permittivity. Then the resonator will form the reference element for a microwave loop oscillator whose oscillation frequency may be counted with high accuracy (for more detail see section 3.3.2). Fig. 4 shows a schematic of such a loop oscillator configured as an absorbed dose calorimeter. We believe that, even at modest cryogenic temperatures such as 50K, δT may be made as low as 1 μ K using, for example, a TiO₂ resonator.

3.6.2 Whispering Gallery Microwave Resonator (with absorber insert)

The first method in section 3.6.1 relies on the availability of a low Z material which satisfies the above conditions on permittivity, thermal properties etc. In the event that no suitable material is available an alternative, but related method, may be explored. By measuring the *whispering gallery modes* in cylindrical resonator rather than the conventional, low order modes, the quality factor of these modes is not significantly reduced even if the central region of the resonator is removed and replaced by a much more lossy material. This central region could then be the ionising radiation absorbing element (e.g. graphite) (also see section 3.3.3).

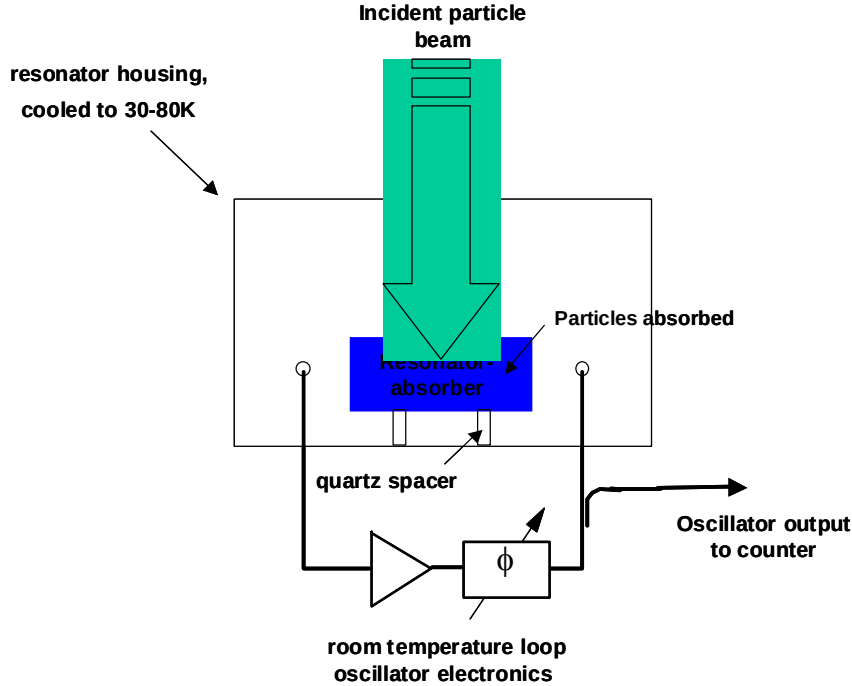


Fig. 4: Schematic of a loop oscillator based microwave resonator thermometer for absorbed dose measurements.

3.6.3 Coupled Dielectric Resonators

Three of the authors of this report are also currently working on yet another novel form of bolometer, based on coupled microwave resonators, which may have widespread application in fields requiring sensitive measurement of the energy absorbed by the impact of particles, whether they be massive particles, photons or even phonons. The detector has emerged from work on functional oxide materials carried out over recent years [15]. The basic idea rests on a system of two coupled microwave resonators, one of which acts as a stable reference frequency but the other of which has a strong temperature dependent frequency (in the present prototype this is made of SrTiO_3 (STO) which has a very strong temperature dependence of its permittivity ϵ_r [16]) (see fig. 5). A key property of this resonator is that if the correct mode is chosen the frequency shift of the coupled mode with temperature does not depend on the volume of the temperature dependent resonator, at least to first order. Thus when energy is absorbed by the STO resonator its temperature changes, resulting in a frequency shift in the coupled resonance. Since extremely small frequency shifts are relatively easily detected this temperature sensor can have nK sensitivity. It also possesses other potential advantages over other sensors types. It is essentially non-contacting, in that interrogation of the temperature is done by the microwave field itself, no cables are required to contact the sensor. Secondly the sensor is an insulator. Its thermal capacity is due only to phonon contributions and there is no electronic contribution. At temperatures below 1K the electronic contribution to the specific heat of metals far outweighs the lattice contribution. Thus an insulating sensor of a particular mass can have a much smaller heat capacity than a metal of similar mass at the same temperature so it may have higher resolution than a metallic one or could be operated at a higher temperature with the same resolution.

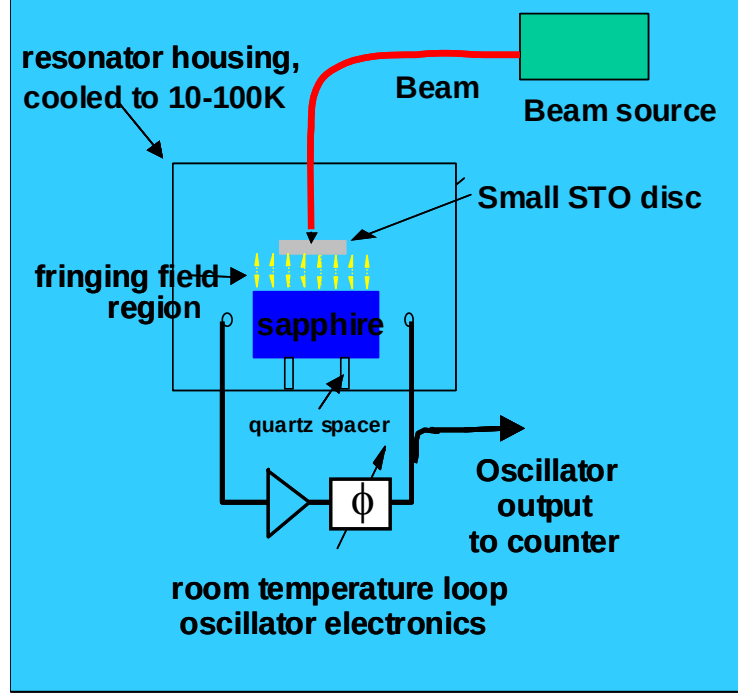


Fig. 5. Coupled microwave dielectric resonators system configured for bolometry/calorimetry.

As an example consider the situation when the microwave fields of a sapphire and STO puck (within the same housing) are only extremely weakly coupled. Then the resonant frequency of a particular mode of the sapphire puck f_0 (assumed temperature independent to a first approximation) is quite unaffected by the STO except for a temperature selected resonance condition when the resonant frequency $g_0(T)$ of a mode in the STO comes into close coincidence with that of the selected sapphire mode. In this situation there is a measurable interaction between the two modes so that they may be treated as coupled independent resonators [17]. The true time independent eigenfunctions are now represented as symmetric and antisymmetric linear combinations of the two unperturbed eigenfunctions. However, within the limits of perturbation theory the following four equations describe the resulting temperature dependent frequencies $f(T)$ and $g(T)$ and linewidths $W_{sap}(T)$ and $W_{STO}(T)$ of the observed coupled resonances:

$$f(T) = f_0 + \text{Re} \left[\frac{A}{(f_0 - g(T)) + iW_{STO}} \right]$$

$$g(T) = g_0(T) + \text{Re} \left[\frac{A}{(g_0(T) - f_0) + iW_{sap}} \right]$$

$$W_{sap}(T) = W_{sap} + \text{Im} \left[\frac{A}{(f_0 - g(T)) + iW_{STO}} \right]$$

$$W_{STO}(T) = W_{STO} + \text{Im} \left[\frac{A}{(g_0(T) - f_0) + iW_{sap}} \right]$$

Here A , B are the normalised coupling strengths between the two modes, proportional to the overlap of the electromagnetic standing wave patterns of the stored energy of the two field distributions of the modes, integrated throughout the housing. W_{sap} and W_{STO} are the unperturbed linewidths of the sapphire and STO resonances respectively. Which of the two coupled modes is observed in any experiment depends on the nature of the input and output coupling structures, especially their positions. If these are situated closer to the sapphire puck then the mode observed is the one which has the dominant stored energy within the sapphire puck. Explicit forms for the coupling coefficients A and B may be written as follows:

$$A = \frac{f_0^2 \int_{vol} \mathbf{E}_{sap} \cdot \mathbf{E}_{STO} d\tau}{\int_{vol} \mathbf{E}_{sap}^2 d\tau}$$

$$B = \frac{g^2(T) \int_{vol} \mathbf{E}_{sap} \cdot \mathbf{E}_{STO} d\tau}{\int_{vol} \mathbf{E}_{STO}^2 d\tau}$$

where E_{sap} and E_{STO} are the electric field vectors for the relevant modes in sapphire and STO respectively. We can differentiate the first of our coupled resonator equations to determine how to maximise the response of $f(T)$ to a change in STO temperature. It is simply shown that the maximum sensitivity to frequency occurs when the resonances of STO and sapphire exactly coincide when

$$\left. \frac{df(T)}{dT} \right|_{\max} = \left[\frac{A}{(W_{STO})^2} \frac{dg(T)}{dT} \right]$$

The first term on the right is the ‘amplification factor’ by which the sensitivity of the coupled system is enhanced over the frequency variation with T of the frequency of the STO resonator alone ($dg(T)/dT$). Thus it is important to operate with coupling between the resonators which is as strong as possible, with the loss tangent ($\tan\delta$) of the STO as low as possible (to minimise W_{STO}).

To demonstrate the potential of the coupled resonator method we have used STO and sapphire pucks (12 mm diameter) spaced by ~ 8 mm in the axial direction. The resonant frequency of the TE_{011} mode in the sapphire puck has been measured as a function of temperature over a wide range (from 20 K to 80 K). Fig. 6 illustrates how the frequency and linewidth of a sapphire resonance changes with temperature as two separate resonances in the STO component come successively into co-incidence with the sapphire mode. We have reported elsewhere [18] that the variation of both frequency and width of a coupled resonance can be quantitatively fitted by the above equations and that the rate of change of resonant frequency with temperature $df(T)/dT$ can be as high as 75 MHz/K. Since the output frequency of a microwave loop oscillator based on a modestly high Q dielectric resonator can be stable to at least 1 in 10^{11} for an

averaging time of 1s [19] the thermometer has a potential resolution of at least 1.5 nK, comparable to low temperature superconducting transition edge thermometers but operating a factor of 10 higher in temperature. The latter would require to be separately temperature compensated using, for example a combination of sapphire and rutile elements [20], as described in the following section.

For operating at cryogenic temperature several cooling facilities already exist within the QDG which could be suitable for radiation calorimetry. For example using liquid nitrogen, or liquid helium cryostats, closed cycle cryo-coolers etc. Fig. 7 shows our coupled microwave resonators system (or other microwave resonators) installed inside a two-stage Cryomech pulse tube cooler, which has a base temperature of ~ 2.8 K, and can provide large cooling power, lower vibration and short distance from room to low temperature surfaces. It is equipped with microwave coaxial lines and optical fibres.

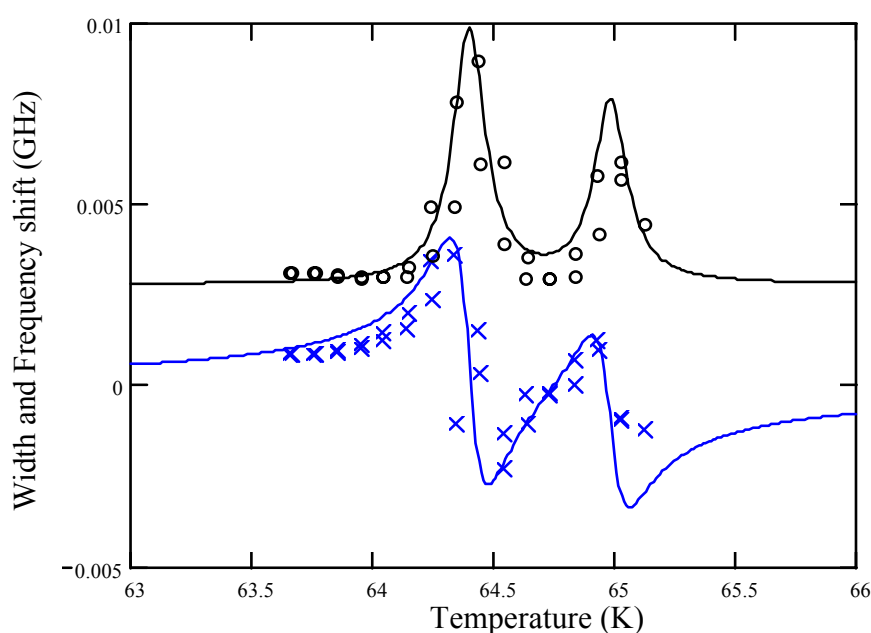


Fig. 6 Experimental results for the temperature variation of a coupled mode (predominantly the TE011 mode in sapphire) as the temperature is changed over a small range. The solid curves represent fits to the experimental data for frequency shift (bottom) and linewidth (top).

3.6.4 High Q Mechanical Resonator Method

A fourth possible method relies on using a quartz resonator both as an absorber and as a high frequency *acoustic* resonator. Certain crystal cuts in quartz result in strongly temperature dependent mechanical resonant frequencies with high mechanical Q . This seems like an obvious approach to carrying out cryogenic calorimetry provided that quartz is regarded as a suitably low Z material. Other materials such as Si_3N_4 or BeO might also be appropriate and would have lower effective Z values. A possible advantage of the mechanical resonance methods is that they may be extendible where appropriate to the very small length scales (for easily absorbed fluences) as exemplified by MEMS devices.

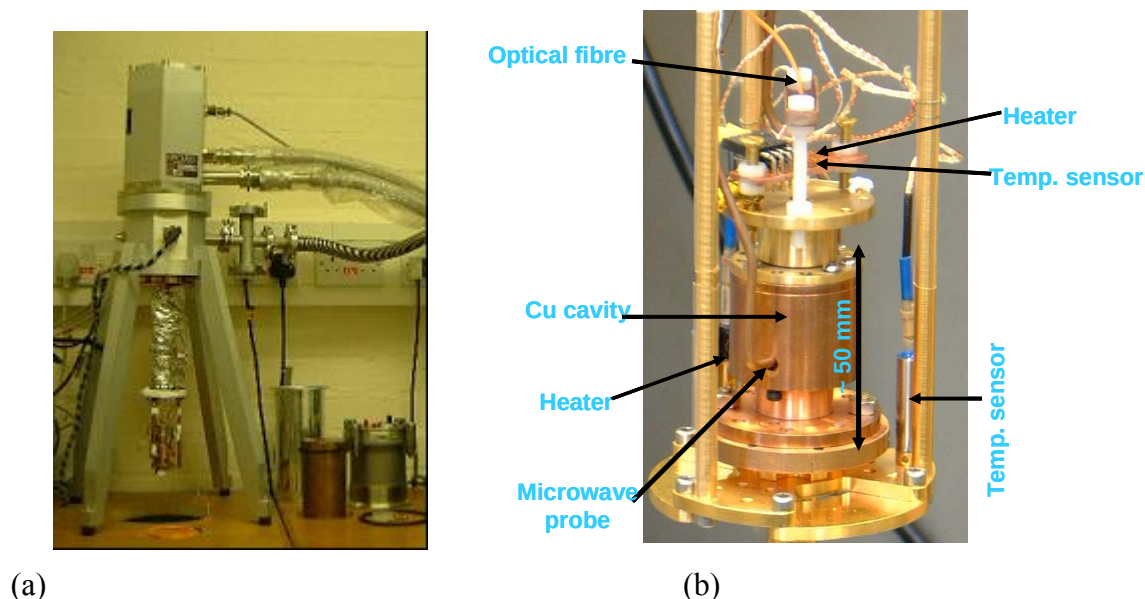


Fig. 7 Coupled microwave resonator system installed in Pulse Tube Cooler. (a) Whole assembly. (b) Close-up of the sensor region: the sensor is contained in a copper cavity for protection.

3.6.5 Magnetic Susceptibility Thermometer

A fifth method could involve the measurement of the magnetic susceptibility of an absorber since with an appropriate choice of paramagnetic centres such a susceptibility thermometer can also provide μK sensitivity. Here it might be possible to dilute a paramagnetic salt in water (ice) or some other low Z liquid such as a hydrocarbon based solvent, to provide a human tissue analogue.

3.7 Ionising Radiation Considerations

A crucial first step in any experimental investigation is to decide on a combination of particle energy, type and fluence which would allow a demonstrator system to be designed, built and evaluated. As a first step before embarking on any experimental investigation a literature search should be undertaken, to understand related existing work and also to tabulate the known properties of appropriate materials for resonator thermometer and absorber functions. We have listed some of these above but there are undoubtedly others which we should investigate. A table of potentially important physical properties is shown below (see section 4).

The mean proton number and the ratio of proton number to atomic mass are important figures of merit in determining the stopping power of any material for radiation of a particular energy and particle type. I represents the ionisation potential. In general low Z materials are preferred for dosimetry applications since they represent a better

approximation to human tissue. The density is also significant from this point of view. The Debye temperature θ determines that temperature below which the specific heat begins to fall rapidly (typically as T^3) and therefore where the temperature sensitivity begins to improve rapidly. The electrical permittivity is important in determining the relative size of the absorber which will have a microwave resonance in a specified range. Thus the larger the permittivity the smaller can be the resonator size. The final column gives an estimate of the how fast temperature uniformity will be established following a changed absorption power within the absorber. The higher the thermal conductivity and the higher the Debye temperature the faster this approach to equilibrium will be.

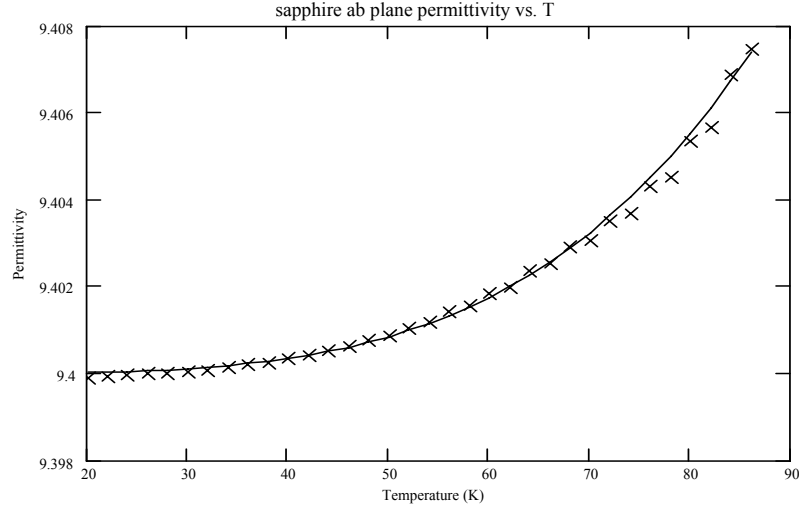
4. Summary of Materials Properties

In this section we summarise some microwave measurements on relevant materials which have already been reported [21]. In addition a literature search has been carried out to determine some thermal properties of these same materials. The proton number and ionisation potentials are also included.

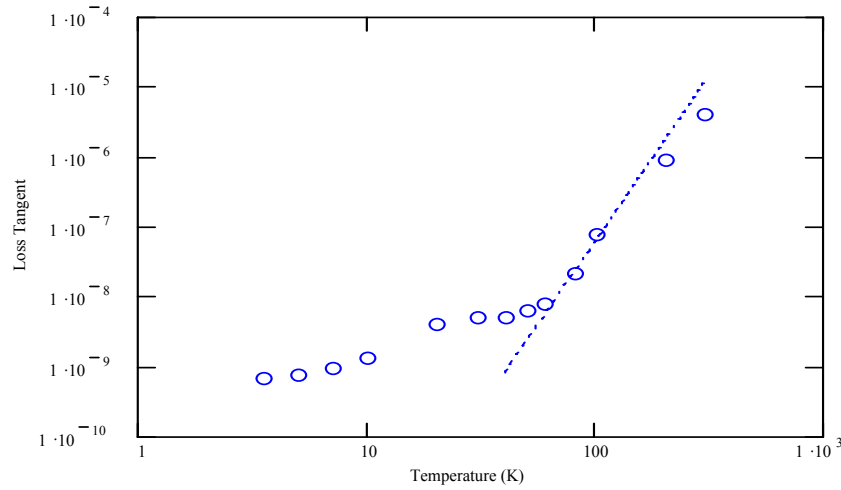
Cryogenics introduces inevitable complications to measurements on dielectrics so that strong incentives are required to make it worthwhile adding this complication. Many properties of single crystal materials show general improvements when their temperature is reduced below ambient temperature, especially the loss tangent and the temperature coefficient of the real component of the permittivity. Single crystal dielectrics have generally lower losses than polycrystalline or amorphous dielectrics at room temperature but this difference is exaggerated as the temperature T falls [22]. For simple ionic materials, with strongly directional covalent bonds and low dislocation densities, such as the single crystal oxides which are widely used as dielectric materials at microwave frequencies, intrinsic loss mechanisms arising from the anharmonic component of the interionic potential, are strongly reduced as T falls. Figure 8a shows results measured in the author's lab of the permittivity of sapphire a function of temperature. Sapphire (Al_2O_3) has a hexagonal structure and consequently the c axis permittivity is different from that measured for the a - b plane by some 20%. The observed losses in sapphire (see fig. 8b) fall with an approximately T^5 dependence, reflecting the change of phonon level occupation numbers, until extrinsic losses begin to dominate [23]. The very low loss-tangent for sapphire observed at cryogenic temperatures (as low as 3×10^{-10} at 1.5K and 3GHz) has led to at least two different application areas, both exploited first by the group at the University of Western Australia. First as the readout resonator for a highly sensitive displacement detector for a resonant bar gravity wave antenna [24] and more recently as the state-of-the-art flywheel oscillator which complements the long-term stability of a cold-atom fountain frequency standard by providing exceptionally low phase-noise for shorter averaging times [25]. Loaded Q values as high as 5×10^9 have been exploited in this latter application.

Titanium dioxide (TiO_2 , rutile) has a considerably higher permittivity than sapphire, between 120 and 80 at room temperature, depending on the crystal axis along which ϵ_r is measured. It possesses two important attributes: the loss tangent is only a small factor greater than that of sapphire, at least down to around 50 K and it has the opposite sign for the temperature coefficient $d\epsilon_r/dT$ [26].

A third general class of dielectrics has extremely high ϵ_r at all temperatures and may enter an ordered, ferroelectric phase as T falls. Thus for example strontium titanate (SrTiO_3) is a perovskite-structured insulator with an exceptionally high relative permittivity (see fig. 9). At low temperatures it enters a paraelectric state before becoming a ferroelectric below about 35 K. Opinions differ as to whether this is a true ferroelectric or a paraelectric whose true transition to an ordered state is frustrated by quantum fluctuations.



(a)



(b)

Fig. 8. NPL measurements of temperature dependent permittivity for a number of single crystal materials. (a) Temperature dependent permittivity ϵ_r for Al_2O_3 . (b) Temperature dependence of loss tangent of sapphire (see [21]). The dashed line is a fit of the higher temperature data to a $T^{4.75}$ power dependence.

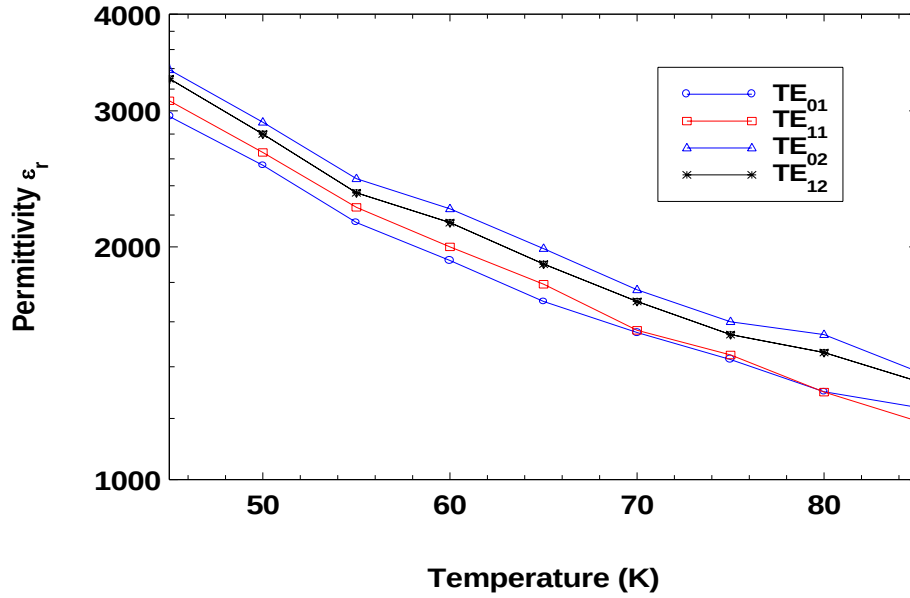


Fig. 9 NPL measurements of temperature dependent permittivity ϵ_r for SrTiO_3 substrate ($10 \times 10 \times 0.5 \text{ mm}^3$), metallised top and bottom, measured using a parallel plate geometry.

The following table (Table 1) lists some relevant properties of high quality single crystal dielectrics which may be suitable for microwave calorimetry. The literature search is not complete at this stage though the gaps in the Table indicate where no reliable information is currently available.

Table 1: Relevant properties of high quality single crystal dielectrics

Material	Crystal structure	$\epsilon_l(300\text{K})$	$\epsilon_l(70\text{K})$	$\delta(300\text{K})$	$\delta(70\text{K})$	$\kappa(100\text{K})$ W/m.K
SiO_2	$\perp$	4.43	4.5	4×10^{-5}		40
	//	4.63				
Al_2O_3	Hexagonal $\perp$	9.4			$< 10^{-7}$	300
	//	11.6				
MgO	cubic	10.4	10.2		$\sim 10^{-5}$	300
LaAlO_3	cubic		24		7.6×10^{-6}	
TiO_2	$\perp$	86	105		$\sim 3 \times 10^{-6}$	
	//	170	230			
SrTiO_3	cubic	1100	1800		5×10^{-4}	

In Table 2 we consider some physical properties of some other materials which may be relevant to future ionising radiation calorimetry studies by microwave techniques. Again the data in the literature is incomplete.

Table 2 Relevant properties of other materials for ionising radiation calorimetry

Materials	Mean Z	Z/A	I	Density ($\times 10^3$ kg/m ³)	Debye Θ (K)	Permittivity at RT	Permittivity at 4K	Thermal conductivity W/m.K
SiO ₂	30.0	.499	139.2	2.32	552	4.5		40
TiO ₂	38.0	.467	179.5	4.26	742	See above Table		25
SrTiO ₃	76.0	.444	249	4.1-4.9	1000	1200	2×10^4	12
BeO	12.0	.486	93.2	3.01	1200	6.7	~ 6.0	290
BN	12.0	.486	83.6	3.487	400		~ 2.4	'high'
Diamond	6.0	.500	78	3.52	2200	5.6		~ 1000
Water (ice)	10.0	.555	95	1.00	192	f dependent		

5. Expected Performance from Microwave Calorimeter

In this section we estimate the likely sensitivity of a dielectric calorimeter based on microwave resonance. Since this is a generic design, rather than one based on a specific application, it is not possible to be too specific since the beam diameter, fluence, absorption coefficients and so on cannot be specified. Instead we will work on the basis that room temperature calorimeters for dosimetry applications require detecting a change of around 10 μ K with a signal to noise ratio of around unity. This is the level achieved with present day graphite calorimeters operated in thermostatic mode for signal averaging times of about 30s [27]. For lower dose rate application we expect a need for increasing the signal to noise ratio by a factor of 100 to 1000. We will estimate how this compares with what could be achieved with microwave resonator calorimeters at room temperature and also at cryogenic temperatures. We base these calculations on an assumed microwave resonant frequency of 10 GHz, with Q values at room temperature of around 10^4 and at cryogenic temperatures (4.2 K assumed for simplicity) of 10^6 . Then we further assume that it is possible to determine the centre of the Lorentzian lineshape to typically 1 part in 10^5 of its width (this is based on our previous measurements of similar microwave resonator based frequency standards).

The minimum detectable frequency shift δf is given by:

$$\delta f = f_0 \cdot \frac{10^{-5}}{Q}$$

And finally the minimum detectable change in temperature ΔT becomes

$$\Delta T = \frac{\delta f}{(df / dT)}$$

The fractional rate of change of resonant frequency $(1/f)df/dT$ of a simple rutile resonator with temperature is around $7 \times 10^{-4}/K$. At cryogenic temperatures this is rather

lower, $\sim 4 \times 10^{-4}/\text{K}$. However the heat capacity is much smaller at cryogenic temperatures so the temperature sensitivity becomes much better.

Table 3 Performance of microwave rutile resonator as calorimeter at 300K and 4.2K

Type of resonator	Temperature (K)	$(1/f)df/dT$	Heat capacity (J/K)	Minimum detectable ΔT change (K)	Thermal time constant	Minimum detectable energy (J/s)
Microwave rutile resonator	300	7×10^{-4}	2.1×10^{-3}	1.4 μK	1 s	3.0 nJ/s
	4.2	4×10^{-4}	5.8×10^{-8}	25 nK	4 ms	1.5 fJ/s

Table 3 shows that already at room temperature a simple microwave rutile resonator has a potentially better performance which is further enhanced by a factor 10^6 at 4.2 K. Note the results in table 3 are calculated for a simple rutile resonator with a diameter of 3 mm and a thickness of 1 mm. Before these results can be transferred to a realistic calorimeter it is necessary to consider the particular application in mind. Thus the beam energy and diameter, stopping length of the resonator material, influence of any vacuum windows required and any possible radiation damage to calorimeter, support structure and windows will all need to be considered. However the very high temperature resolution and energy sensitivity make it clear that the microwave resonance method has many possible advantages.

6. Future benefits of the work to NPL

In this brief section we summarise some advantages which would be brought to NPL through the development of microwave resonance based calorimetry.

- **Innovative application of cryogenic technology to dosimetry standards**
- **Development of more sensitive calorimeters**
- **Development of a microdosimetric calorimeter**

These will be designed to meet the requirements for internal sources and low and medium energy x-rays and play a pioneering role in this field. In addition this work would improve prospects for funding of a collaborative project with e.g. McGill University for the development of brachytherapy calorimeters, as well as NMS funding of next generation absorbed dose standards.

To develop a microdosimetric calorimeter would yield a unique instrument for the independent measurement of microdosimetric quantities. Application to low-energy photon and electron sources as well as to light-ion beams would substantially contribute to the understanding of biological effects of radiation.

In the QDG we also have developed a nanoSQUID based bolometer at cryogenic temperature which is capable of the highest sensitivity [28]. Although it is not the subject of this report, this SQUID-bolometer could possibly serve as a micro- or nano-dosimetric detector.

7. Conclusions

The report has considered in some detail the likely use to Radiation Dosimetry (RD) of the development of various microwave resonance based calorimeter designs for dosimetry applications. The predicted sensitivity of microwave resonance calorimetry makes it clear that such a system is competitive with conventional calorimetry, even at room temperature. Much greater advantages accrue if cryogenic operation can be implemented in an ionising radiation application. In addition a number of other possible longer term applications of these technologies have also been outlined which should be considered for future collaborative work between DEM and DQL scientists.

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