A DISSERTATION SUBMITTED FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

An Inductive Superconducting Transition-Edge nano-Detector for nano-Dosimetry Applications

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Summary

In this thesis I have demonstrated the feasibility of using Inductive Superconducting Transition-Edge Sensor (ISTED) as an excellent nanodosimeter for nano-dosimetry applications. This thesis can be divided broadly into two parts. In the first part, I begin by addressing how the study of low energetic ionising particles in liquid water using concepts from classical mechanics is a valid approach, despite being inside the quantum-classical boundary regime. Based on the circumstantial validity condition, I showed that the percentage uncertainties in nanodosimetric quantities due to Heisenberg’s uncertainty principle for sub-1 keV electrons in liquid water as calculated by GEANT4-DNA is not significant enough to cause changes to their distributions. Important nanodosimetric quantities studied in details are ionisation cluster-size distribution, second order of moment for cluster-size distribution (M₂) and the cumulative frequency of ionisation cluster-size distribution from cluster-size two (F₂). In the second part of my thesis, I have focussed on the design, optimisation, fabrication, characterisation of the superconducting devices. An ISTED is made from three components: a) a Superconducting Quantum Interference Device (SQUID), b) a superconducting thin-film and c) a top layer of thin-film Carbon absorber. I have measured the magnetic flux noise of a nanoSQUID of loop dimension 350 nm and nano-Josephson junctions of dimensions 65 nm x 65 nm as $3 \times 10^{-14}$Φ₀² in the white noise region. It is shown that the measured nanoSQUID is more than capable of sub-10 eV energy detection.
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Chapter 1

Introduction

In 1896, a German physicist Wilhem Röntgen made a remarkable discovery of x-rays radiations. Since the discovery of radiations there have been revolutionary advances in non-surgical radiotherapy treatments. Radiation is defined as the emission or the transmission of electromagnetic waves or subatomic atoms, which causes ionisations in a medium. Shortly after this discovery, medical doctors such as Emil Grubb (Chicago, USA) pioneered the new possibility of utilising radiations as a mode of non-surgical medical treatments. Today, radiotherapy is still considered the largest non-invasive treatment modality used in cancer treatments. Accurate characterisation of the effects of such radiations on human tissues is an important research area to ensure the most effective delivery of radiation with minimal side effects. The quantification of the effects of radiation in biological systems is known as the field of radiobiology. In parallel with developments in medical radiation physics, the research of bolometry and calorimetry has also seen rapid advances. The purpose of this project is to understand dosimetry at the nanoscale using novel nano-detectors with consideration of their future use in radiobiology.
1.1 Project context

The proposed project aims to further improve current experimental dosimetry at the nano-scale using sophisticated and low noise cryogenic detection devices and methods. The nanocalorimeter of focus is an Inductive Superconducting Transition Edge Detector (ISTED) first developed at the National Physical Laboratory [65]. The proposed experimental set-up is designed to measure the energy deposition with nanometre cross section resolution, typically of 300 nm. The sensitivity of the device is proposed to enable the measurements on absorbed energy distributions, which may provide the radiation metrology community a new or supplementary nanodosimetric quantity to quantify radiation quality, alongside the conventionally used ion cluster size distribution.

Given the large variation in the possible studies involved in nanodosimetry it is essentially a multidisciplinary area of research, consisting of collaborations between biologists, chemists, physicists and computer engineers. The long term and overall goals of this project are tied in with the European Metrology Research Programme (EMRP)- Biologically weighted Quantities in RadioTherapy (BioQuaRT)[126], where three main areas of research associated with medical radiation metrology may be identified as follows:

1. To work towards the nanoscale metrological definition of a radiation quantity which may infer radiobiological effectiveness.
2. To work towards developments on appropriate experimental realisations of the above.
3. To validate experimental methods through appropriate simulations.

The research goal of the author’s PhD project is to demonstrate relevant and plausible nanodosimetric experimental approaches at National Physical Laboratory (NPL) and University of Surrey (UoS).
1.2 Novelty of project

Distinct improvements and differences in the proposed nanodosimeters to the existing ones are summarised as follows:

- Nanodosimeters using superconductors as sensitive energy absorbers with nanometre depth profile can be realised.

- The nm-pixel resolution on the absorbed energy distribution can provide invaluable information for the ionising radiation metrology community, as it may be used in conjunction with, or even substitute the current commonly used ion cluster size distribution as a better nanodosimetric radiation quality.

- The use of cryogenic detectors as opposed to room temperature electronics reduces the associated thermal fluctuations in the electronics.

- The ISTED absorber is a passive material, which means that the sensitivity is independent on any electrical connections.

- The flux coupling between the absorber and the SQUID means that the read-out SQUID does not suffer from dissipative losses due to electrical connections.

1.3 Microdosimetry and nanodosimetry at NPL and UoS

Collaborative works on microdosimetry between UoS and NPL initiated in 2009 [49] and was coordinated by Prof Ling Hao (NPL), Dr Hugo Palmans (NPL), Prof Andrew Nisbet (UoS) and Prof Karen Kirkby (UoS). The Niobium films used for most of the devices presented in this thesis were mainly fabricated by the University of Strathclyde and Physikalisch-Technische Bundesanstalt, Germany (PTB). Devices were milled by Dr David Cox, typically using Focussed
Ion Beams (FIB) at the Surrey Ion Beam Centre. This was an interesting and novel departure from the commonly used gas interaction chambers employed by other groups, with reasons described later. Our research group specialises in the use of small-scale cryogenic calorimeters to study dosimetry on the micro- and nano-scales.

The first realisation of the concept for ISTED and preliminary results were conducted by Prof Ling Hao and Prof John Gallop in 2003 [65, 63, 64]. The current microISTED cryostat was constructed by Dr Sebastian Galer, and Dr. Kamran Faithi (was a PhD student) focussed his research on the modelling aspect of the thermal transport and the response mechanisms of the detector to medical radiations using Monte Carlo methods [45]. In conjunction to the development in microdosimetry, our group has recently extended research interest into nanodosimetry, as the PhD project for the author. The justification for the reduced spacial investigation to nano-scale is described in chapter 2.6. A list of publications, oral and poster presentations arising from this PhD project are listed in Appendix A.
Chapter 2

Microdosimetry and nanodosimetry basics

Radiotherapy is the most common non-invasive treatment method used in the treatment of cancer, thus improving the effectiveness of such treatments is of great importance. Minimising the damage to healthy cells whilst maximising the damage to targeted and cancerous cells are the common goals for all medical physicists working in the field in radiotherapy.

There are some important length scales in a biological system. Starting from the size of a typical tumour, approximately 3 cm in diameter, to the size of each cell that is approximately 100 µm in diameter. Twenty times smaller than the cell, the cell nucleus is roughly 5 µm in diameter. As we probe deeper into a nucleus, each chromosome fibre is 200 times smaller than the nucleus at 25 nm in diameter. At the shortest length scale of interest, the chromosome fibres are made from twisted strands of DNA, which are 2.3 nm in diameter. The accepted view on effective cell destruction is associated with the damages to DNA strands. This is because it is the most vital component of a cell as it encodes information for almost all cell functions.
Radiobiology is a research area that can be loosely described as the determination of dose-effect responses from an ionisation radiation to biological systems. Absorbed dose is an appropriate quantity for the description of high-level exposures that are more likely to involve multi-cells interactions. However, as the length scale of study approaches single cell or even further to DNA structures, such as when studying interactions of low energy ionising particles, the use of macroscopic quantities such as absorbed dose becomes unsuitable for these single cell effects. The problems associated with using dose is amplified through the determinations of relative biological effectiveness (RBE). Given the definition of RBE, it is implied that RBE is a combined measurement of two factors: first, the severity of the effect, and second, the probability of the effect. However, as will discussed in detail later that severity is not appropriately characterised. Instead of dose or RBE, it has been suggested that the correct approach to quantifying radiobiological effects is the concept of hit-size effectiveness function (HSEF). HSEF relates the absorption of energy in a more meaningful way than dose. It relates lineal energy density to the probability distribution of a biological response. HSEF and together with hit-size distribution is said to be replacing RBE and dose at the microscopic view.

2.1 Radiation interaction with matter

The fundamental concepts in radiation physics or dosimetry can be found in numerous textbooks and relevant publications by the International Commission on Radiation Units and Measurements (ICRU). In this section, a selection of the important concepts will be introduced.

The effects of ionising radiation on matter depend upon the amount of energy deposited within the target. This is based on the assumptions that:

- The deposited energy is spread evenly through space.
- The radiation related damage to the medium is proportional to the absorbed dose.
The concept of absorbed dose, \( D \), defined in Eq. 2.1 is typically expressed in units of J/Kg or Gy, where 1 J/Kg = 1 Gy.

\[
D = \frac{\Delta E}{\Delta m}
\]  

(2.1)

The definition of absorbed dose is not enough to quantify the radiobiological effects in living cells. An irradiation experiment on the survival fractions (SF) of the CHO-K1 Chinese hamster cells under exposures to various ionising radiations has shown that the absorbed dose, \( D \), is not an absolute indicator in relation to the survival fractions of those cells [78]. To describe the radiobiological effects of ionising particles- Relative Biological Effectiveness (RBE) is employed. It is defined as the ratio between a reference dose, typically chosen as photons, to a dose of any other chosen particle of interest to produce the same level of biological damage, Eq. 2.2.

\[
RBE = \frac{D_{\text{ref}}(SF)}{D(SF)}
\]  

(2.2)

Clinically, the use of the RBE value is dependent on a number of factors, such as the irradiated particle type, energy and the given dose, and the targeted cell or tissue types [74]. Also of relevance is the Linear Energy Transfer (LET), or also known as the stopping power. This is defined as the depth dependent energy transfer (to the medium) along the path of the incident particle, defined as shown in Eq. 2.3.

\[
LET = \frac{\Delta E}{\Delta x}
\]  

(2.3)

LET describes the energy loss of the incident particles to the surrounding medium, \( \Delta E \), over a transverse distance of \( \Delta x \). However, as the length scale of interest reduces towards the micrometer it becomes more appropriate to measure \( \Delta E \) over a small volume. This is known as
the ‘lineal energy’ and is the focus of studies in microdosimetry. A typical measured volume is equivalent in size to a liquid water sphere with 1 µm diameter. However, the concept of microdosimetry is disadvantaged in two ways. First, the length scale of interest are comparable to the track length, meaning that it suffers from losses of information on the track structures. Second, even at the micrometer scale the length of a single DNA strand is still roughly 5 times smaller, which may be significant enough to make microdosimetry slightly too large for accurate investigations into the biological effects to radiation.

2.2 Heavy charged particles

Interactions between heavy charged particles (HCP), such as α-particles and protons, and matter mainly occurs through Coulomb interactions between the ion’s positive charge and the negative charge of orbital electrons from atoms in target medium. The likelihood of interactions of the heavy ion with the target’s nuclei is relatively much smaller. Possible forms of interactions of the heavy ion with electrons are excitations and ionisations. Excitation occurs when an electron is raised to a higher energy/orbital level as it receives the required energy to do this from the passing charged particle. Ionisation occurs when the electron has received energy greater than its binding energy such that it is completely removed from its original orbiting atom. The energy transfer from the heavy charged particle to the electron causes the charged particle to slow down in the target medium. The charged particle continues to interact with the medium until it loses all its kinetic energy and comes to a stop. When the ion becomes less energetic its trajectory tends to be fairly straight as further interactions with electrons do not cause much deflections given the huge difference in mass between the ion and electrons. It is for this reason that the trajectories of charged particles can be characterised by a finite range within a given target medium.
Typical products of interactions between HCP and orbital electrons are usually ion pairs of excited atoms and ejected electrons. Recombination between the ion pairs do occur and the result is a neutral atom. In some types of dosimetric detectors such as semiconductor counters the rate of recombination is suppressed as ion pair formation forms the key basis to detector response. Ejected electrons of enough kinetic energy can undergo further ionisation interactions with the target medium. These energetic ejected electrons are known as secondary electrons or $\delta$-electrons, whilst the original incident HCP is known as the primary particle. Energy transferred from secondary particles are called indirect energy transfer as they are not directly from the incident particle. The majority of a primary particle’s energy is lost through creations of secondary electrons. Since the mean free paths of secondary particles are short in comparison to the incident HCP, we see that ionisation from secondary particles are in close proximity to the trajectory of the primary particle.

Sometimes, ejected electrons have enough kinetic energy to create further ionisations. These energetic electrons are called secondary electrons or $\delta$-rays and represent an indirect transfer of energy from the HCP to the material. The majority of HCP energy loss occurs via $\delta$-rays. However, the range of the $\delta$-rays is very small compared to the incident HCP so ionisation occur very close to the HCP track.

### 2.3 Electrons

Ionising particles with low mass and low energy such as secondary electrons with energy lower than 500 eV are particularly damaging towards biological targets. With inelastic mean free path length of only 1-2 nanometers [40, 129] they are the main contributors towards lateral energy deposition from a primary incident particle. Secondary electrons typically have energies below 1 keV and they undergo electromagenetic interactions either inelastically or elastically. In both
cases the electrons can change direction of motion post-interaction, but in the case of inelastic interactions they lose some of their energy in the process whilst exciting one or more electrons in the target atom/molecule to an higher or even continuum energy state, which results in an ionisation of the target atom/molecule. Single ionisation potential depends on the binding energy of the outermost electron valence bound. By convention, the electron of lower energy, typically of $E \leq (T - B_i)/2$, is defined as the secondary electron due to the indistinguishable nature of the two electrons. In elastic interactions no energy is transferred, where $T$ represents the energy of the incident electron and $B_i$ is the binding energy of the $i$-th energy level of the target atom/molecule. For electrons of energies less than several eV, a process called resonant electron-impact may also be an important contributor towards biological DNA damage [13]. Low energy electrons may result in formations of transient molecular anions during electron attachment processes. The attached electron may auto-detach which may vibrationally excite the molecule or leave behind a dossiciate of the molecule.

### 2.4 Light ions

Light ions are also an important group of particles in the context of inducing significant biological damages [69]. For instance, protons and helium particles of energies between 100 keV and 20 MeV will fall into this group. Like other non-relativistic charged particles, they also interact with matter via electromagnetic processes such as ionisation, excitation and charge-transfer. Elastic collisions have a small scattering angle profiles and are of low probability, and thus they are usually neglected [91]. Similarly, charge-transfer interactions that results in formation of anions are also of low cross section for the energy range of interest, which means it is of low probability and is neglected [124]. Light ions with greater energies than 20 MeV are typically not studied for their radiobiological impact as their free mean path length are large relative to
the length scales of a DNA segment.

2.5 Projectile with varying velocities

Another way of categorising ionising particles is to consider its velocity with respect to the average velocity of valence electrons in the target medium. When the velocity of the incident particle is much greater than the velocity of the valence electrons, typically for electrons and light ions this is above 500 eV and 1 MeV/u respectively, the incident particle is known as a fast projectile and they cause minimum disturbance in the electron distribution of the target atom/molecule. Since the time window for transfer of momentum during interaction is short in comparison to the orbital frequency of the target electron, it is applicable to use the static-field approximations. This means that the charge distribution of the target can be approximated as being spatially static and only very small energies are transferred to the target during the interaction (scattering process). First Born approximation is a first order perturbation theory that can be applied to the interaction between incident fast ions and target matter, where the treatment takes the incident particle as plane waves [8, 73]. For instance, elastic collisions under the first Born treatment is described as scattering of the incident plane wave by a central static potential $V(r)$. The central static potential is an electron density distribution profile of the target atom/molecule [32, 96]. When the de Broglie wavelength of the incident ionising particle is much smaller than the distance between the target atoms, the scattering process can be described purely from the potential of a single atom, known as the independent atom model (IAM). However, the most dominating interaction for a fast projectile is inelastic processes, namely ionisation. Generalised oscillator strength [46] can be used to described these inelastic interactions, and it is assumed that the interaction can be further approximated by optical oscillator strength due to its dipole characteristic. As the name suggests, the optical oscillator
strength is related to the absorption cross section of photons, and is a measure of the transition probabilities within the target molecule approaching zero momentum transfer.

As discussed earlier, ionisation interactions dominated the fast projectile situation, however as the projectile loses its energy and enter the ‘slow’ regime, interactions such as electronic, vibrational, charge transfer and rotational excitation become more dominating. For instance, electrons of energies of several eV the dominating interactions are electronic, vibrational and rotational excitations. The likelihood of rotational excitations is positively related to the dipole moment of the target molecule, i.e. the higher the dipole moment the higher the cross section or likelihood of occurrence it has.

Unlike fast projectiles, the trajectory path of slow projectiles usually shows large angle deflections as they are likely to transfer a more significant amount of its momentum to the target molecule/atom during interactions. For elastic collisions, the incident particle remains to keep its original energy but undergoes changes to both its energy and momentum in inelastic collisions. The spherical complex optical potential (SCOP) model can be used for the target potential in describing elastic scattering of electrons of around 100 eV. Here, the scattering potential is assumed to be approximated by the optical potential \( V_{\text{opt}}(r) \) \[32, 36\], as shown in Eq. 2.4.

\[
V_{\text{opt}}(r) = V_{st}(r) + V_{\text{exch}}(r) + V_{\text{pol}}(r) - iV_{\text{abs}}(r)
\]  

(2.4)

where \( r \) is the distance from the centre of mass of the target. \( V_{st}(r) \) represent short-range potential, which is assumed to be static and spherically symmetric during collision. Exchange potential, \( V_{\text{exch}}(r) \), accounts for the exchange interactions between the incident and target electron. Since the two interacting electrons are indistinguishable and it is important to consider possible interchange between them, especially so when the velocities of the two particles are similar \[37\]. \( V_{\text{pol}}(r) \) is the correlation-polarisation potential, which accounts for the interference of the incident particle wave and the electron potential of the target and so it regulates
the probability profiles of scattering [42, 46, 32, 133]. An imaginary potential, $V_{\text{abs}}$, describes the energy absorbed through means of inelastic interactions. The greater the energy of the incident particle the greater energy contribution it is towards inelastic interactions [32]. The scattering potential given above are for atomic configuration, but if the target is a molecule of multi-center potentials then a correction treatment would be to simply take into account of the center of mass for the molecule as well as intra-molecular scattering [37, 32] across different atoms. This treatment is more appropriate for low energy electrons (below a few tens of eV) as the de Broglie wavelength of low energy particles can stretch across several atoms [53]. Electrons of energies below 500 eV are subject to higher-order perturbation effects in their inelastic collisions. Electron-capture interactions by the incident particle on a bound target electron occurs with increasing probability and peaks for when projectile velocity is similar to that of the bound electron [133]. This charge-transfer process is important in the low energy range as the incident electron ionises the target molecule by removing a bound electron. It is far more likely for an ion with higher charge to undergo ionisation interaction than one with less charges for the same medium. This is because the captured electrons can act to screen the electrical potential of the incident ionising particle [91, 133]. Electrons can, in turn, be lost from the projectile, leaving one or more free electrons behind. After a few interactions, it may be that a charge equilibrium of an incident ion beam is reached. This means that rate of forming and dissociation of the certain charged particle reaches a steady-state.

### 2.6 Nanodosimetry

One end goal of dosimetry studies in general is to improve risk estimates of human exposure to ionising particles, and in particular to low-LET radiation, due to diagnostic, occupational or environmental reasons. The complex interaction mechanism of radiation to biological entities
involves a huge spatial range from DNA strands to the biological entity as a whole. However, initial events are best described physically. Together with better understanding of biological mechanisms it will be easier to start from a host of initial events to how it eventually leads to onsets of cancer.

Microscopic dosimetric quantities such as absorbed dose which is calculated from the stopping power and is linked to the average energy lost by the incident particle. However it does not characterise molecular changes as molecular changes within a given microscopic target volume depends on the energy deposited or similarly related to the number of interactions [121]. The difference between the absorbed and deposited energy with respect to the target medium and incident particle respectively is due to the extra energy transported away from the microscopic target volume by energetic secondary electrons. Secondary electrons with high energies are capable of transferring their energies over a significantly greater distance than the length-scale of the target volume. The number of ionisation events in a macroscopic target is seen to be related to the energy deposited by the average ion pair production energy. The average energy for the production of an ion pair is called the \( W \)-value. However, whilst the approximation works well when the number of ionisation events is larger, the relationship breaks down when the target volume is small.

It follows that the concept of absorbed dose is very limited to nanodosimetry and should be replaced with concepts from detailed particle track structure, where it follows a probabilistic density distribution.

There are two common ways of studying the relationship between particle track structure and radiation induced damage to target volumes of a few nanometres in dimension. The first approach is by starting from the energy deposition in the nanometric volume and relating this to the initial physical damage to DNA [109]. However, as energy deposition is not a direct measurement an easier measureable quantity is the overall charge of particles produced dur-
ing ionisation, and it can be translated into energy deposition using the W-value, as mentioned above. Nevertheless, the use of this conversion to energy deposition is not applicable to volumes of nanometer dimensions. The second approach is to start from the mean number of inelastic events and use it to estimate an yield of DNA strand breakages [9]. Both of those two approaches incorporates interactions such as excitations into their results, which is very important as excitations can lead to neutral dissociations of important biological molecules such as DNA in the target volume.

More work has been done to overcome the limitations associated with measurements on macroscopic volumes [33, 56, 123, 131]. It has been suggested that nanodosimetric experiments that utilises a low-density gas volume can be used to measure track structure related quantities. These experiments can be validated through track structure simulations that often are computed using Monte Carlo methods. After validations, the simulation can even be adjusted from simulations in gas-filled volumes to condensed matter [33, 58, 123].

The concept of experimental nanodosimetry was first proposed as early as the 1970s at the 4th and 5th Symposia on Microdosimetry [122]. Early works on understanding the mechanisms of biological damages began as Monte Carlo simulations of radiation transport of ionising particles such as electrons [118, 110, 112, 106] and light ions [118, 147, 140, 23]. However, experimental nanodosimetry did not begin until about 1995 [15]. It was a challenge to the metrology of ionising radiation at the time, and remains so today. It is not clear what are the key parameters of an ionising radiation that determines its radiobiological effectiveness. However, in order to characterise the radiation effectively the parameters of interest should: a) correlate closely with the track structure of the ionising particles, and b) be able to resolve to the nanoscale of particle interactions given the stochastic nature of the interactions. This is based on the assumption that the radiobiological damage to DNA strands can be described by a radiation quality (in other words, its track structure), such as the number of particle interactions or the number of
ionisation events at the nanoscale.

Experimental nanodosimeters are typically designed to measure the cluster sizes of ionisation, \( \nu \). This is a quantity used to describe the number of ionisations produced by a particle in a specific volume. Here, the number of ionisation events is assumed to be the dominating factor in damaging segments of DNA. The probability, \( P_\nu(T) \), of an ion cluster formation with size \( \nu \), as a function of particle energy, \( T \), is a widely used parameter to measure with nanodosimeters. Similarly, the cumulative cluster size probability, defined as the probability for cluster formations with sizes \(< \nu \) is another important definition, see Eq. 2.5.

\[
F_\nu = \sum_0^\infty P_\nu
\]  

(2.5)

When the spatial dimension of interest is comparable to a single strand of DNA, \( P_1 \) and \( P_2 \), the probability for the occurrence of one and two ionisation event respectively, becomes two special cases in view of the DNA strands. It is commonly accepted that the probability of causing one ionisation, \( P_1 \), should correlate with the probability of forming a single strand break (SSB). Similarly, \( P_2 \) is proportional to formation of double strand break (DSB). SSB and DSB are very important parameters in calculations to quantify the radiobiological damage.

The most important nanodosimetric quantity is the ionisation cluster size, which is defined as the number of ionisation events caused by a single incident primary particle in a given nanometric volume [56]. A typical target volume is a cylinder with radius and height that are equivalent to a segment of DNA of 10 base pairs. The commonly used dimension this cylinder in the literature is one with height of 3.4 nm and diameter of 2.3 nm. Since the human body is most abundant of water molecules, the biological sensitive volume is generally defined to be filled with liquid water. Ionisation interaction events occurs with a probability and the likelihood of an event is determined by the ionisation mean free path. In other words, the ionisation cluster size \( \nu \) can be viewed as stochastic at the nanometer length-scale, and with
any probabilistic variables it should be better defined by a probability distribution. Since the ionisation mean free path depends on the radiation quantity $Q$, the probability distribution of ionisation cluster size $P(v|Q)$ also becomes a characteristic of a specific radiation quality $Q$. More track structure related quantities can be derived from the statistical momenta of $P(v|Q)$ by the following:

$$M_k = \sum_{i=0}^{\inf} v^i P(v|Q)$$  \hspace{1cm} (2.6)

Nanodosimetric quantities based on the ionisation cluster size distribution $P(v|Q)$ is seen to explain results from biological experiments fairly well [50, 51, 58, 105]. These findings indicates that the nanodosimetric approach based on the ionisation cluster size distribution is appropriate to estimate biological effects. The cumulative probability, $F_2$, is used as a parameter describing the complexity of clustering, see equation 4.1. The biological rationale behind this parameter is that at least two ionisations of the DNA strands are required to form a double-strand break.

$$F_2 = \sum_{i=2}^{\inf} P(v|Q)$$  \hspace{1cm} (2.7)

Ionisation cluster distribution are frequently used in nanodosimetry. The number of ionisations per incident particle are counted.

Nano-calorimeters are used to study the track structures and energy deposition profiles associated with ionising particles in radiotherapy. Over recent years, various designs on nanocalorimeters have been proposed and experimented with. In section 2.7, we will review some distinct groups of existing nanodosimeters, before moving onto nanometricS cryogenic calorimeters.

### 2.6.1 Monte Carlo track structure simulators

The production and subsequent diffusion of reactive particles during the energetic degradation of secondary electrons has a significant nanoscale effect on the energy deposition distribution.
of radiation. The coupling of the actions of these reactive chemical species to radiation particles is an ongoing effort. A comparison of Monte Carlo codes applied in radiation dosimetry is provided by Nikjoo [114], which is briefly summarised in this section.

Monte Carlo codes for radiation transport can be separated in two main groups: so called condensed history simulations and track structure simulations. Condensed history simulations are useful if the target size is macroscopic. Such simulations combine many single interaction in one step to allow a fast transport calculation of adequate accuracy. Many of these codes use the stopping power to determine the energy loss of the incident particles in the medium and generally multiple scattering theories are applied for electron transport. A list of condensed-history codes together with treated particle types, energy ranges and target media can be found in [114].

For microscopic target volumes such as the DNA, track structure Monte Carlo codes are better suited. Such codes transport a particle interaction-by-interaction and, therefore, provide detailed histories of the particle’s track. The modelling of individual interactions is enabled by the exclusive use of interaction cross section instead of multiple scattering theories and macroscopic quantities, such as the stopping power. The downside of such detailed simulations is that they are time-consuming and therefore limited to microscopic spatial dimensions. Track structure codes have been developed since the 1960s and can be classified in three groups based on the time scale [114]. The first group of codes provide the physical track structure, occurring in the first $10^{-15}$s [117]. Examples are Geant4-DNA [72] and PTra [56]. The subsequent transport of chemical products, produced in the physical stage, is additionally handled in codes such as PARTRAC [48] and KUBRUC [138]. Biokinetic processes, such as DNA damage repair, are additionally treated after the chemical stage, for example, in code PARTRAC. Originally developed in the late 1990s, the detailed track-structure code PARTRAC models the time-dependent interaction of reactive chemical species with DNA material. For a comprehen-
Models of DNA molecules can have different levels of complexity. The complexity ranges from a simplified linear DNA segment, which is modelled in the form of a cylinder filled with water [58] or water vapour [106] to segmented targets of water [149]. Highly structured DNA targets are also available and it uses interaction cross sections of liquid water for radiation transport calculations [11, 48, 108]. Simulations codes using interaction cross sections of DNA constituents also exist for electron transport simulations [35]. The differences between track-structure codes lies primarily in the theoretical models and cross-sections that each codes uses [115]. The agreement of most track structure codes is good at high-energy; however, at lower energies the consensus becomes less clear. The uncertainty of cross-section data for low-energy electrons in water is problematic. Their influence upon the ultimate spatial distribution of energy deposition is certainly significant. Existing cross-sections for low-energy interaction in liquid water are derived from water vapour experiments. The effect of phase difference on yield is significant. The theoretical assumptions made by the authors of each code in deriving these cross-sections have appreciable impact upon the nanoscale action of the codes [139]. The development of the GEANT4-DNA toolkit [71] represents the evolution of low-energy Monte Carlo simulation. GEANT4’s design and versatility brings these simulations into the mainstream.

### 2.7 Current nano-dosimetric experimental techniques

A number of detectors are based on gas chambers. Most of these gas chambers are designed to detect direct ionisations caused by the primary radiation. In this section I shall examine a selection of the currently available nanodosimetric detectors.
2.7.1 Ionisation chamber counters

The work on nanodosimeters initiated as early as 1996. At the 12th Symposium on Microdosimetry in Oxford, the design of an ion-counting nanodosimeter and characterisations on the ion transport in low-pressure propane gas was presented by the Weizmann Institute of Science, Israel [132]. Further work on the next generation nanodosimeters began with collaborations from Loma Linda University (The Department of Radiation Medicine), Weizmann Institute (Detector Physics Group) and the University of California, Santa Cruz (Santa Cruz Institute of Particle Physics). The joint work resulted in the development of two complete systems of a type of ion-counting calorimeter with silicon tracking telescopes, of spatial resolutions equivalent to the nanoscales. These systems are still used in activate studies at present; one of the two systems made stayed at Loma Linda University, whilst the other was located to Physikalisch Technische Bundesanstalt (PTB) in Germany.

Ionisation chambers rely on the creation of ion pairs to form an electrical signal. Hence, the total number of ion pairs created along the radiation track is of interest. However, the incident particle may also lose energy via interactions that do not produce ion pairs, such as excitation. Collisions between positive ions and free electrons may result in a process called combination. This is where the electron is captured by the ion which makes them a neutral atom together. The positive ion may also collide with a negative ion causing the extra electron to be captured by the positive ion making both ions and neutral atoms. In both cases, the charge in the original pair is lost, and thus does not contribute towards the output signal. Recombination can be suppressed by the application of an external electric field, which moves the charges away from their point of origin under drift. Positive ions move in the direction of the conventional electric field and negative ions move in the opposite direction. The drift of positive and negative charges constitutes an electric current. If the detector is at a steady-state irradiation, the rate of formation of ion pairs will be constant and exactly balanced by
Figure 2.1: Diagrammatic layout of a typical ion counter. Ionising particles are directed in the direction to a trigger detector inside a gas-filled interaction volume (IV). Any charged interaction products produced along the trajectory of the incident ionising particles are collected within a candle-like volume called the sensitive volume (SV). They are then accelerated through a series of charged plates before being detected. Reproduced from [52].
the rate at which charge is lost due to recombination, diffusion or migration from the volume. This means that the measured current is a direct reflection to the rate of ion-pair formations in the interaction chamber, assuming that the detector is 100% efficient and that recombination is a negligible process. Ion chambers are typically operated at voltages in the ion saturation region, where a sufficiently high voltage is applied to create a large enough electric field to suppress recombination, and further increases in voltage do not increase the current as the rate of formation becomes steady.

An ion-counting nanodosimeter consists of a low pressure gas chamber, sets of voltage gating electrodes and an ion counting detector. Along the path of the incident particle the gas chamber is fitted with silicon tracking telescopes at both of its ends, which are used to count the number of primary particles entering and exiting the gas chamber, enabling the verification of the predicted ion-extraction profiles with 0.1 mm spatial resolution. As the primary particle beam interacts with the gas particles in the interaction chamber, the ionic products of such interactions are accelerated and collected towards the ion counter using electric fields maintained by sets of electrodes. The collected column of ionic particles (the sensitive volume) forms a candle-like structure, typically with effective diameter of 1.5 mm. Efficiency of the ion-extraction column is measured as 50%. The gas chosen for the interaction chamber is propane.

Given the gas is maintained at low pressure at 1.33 mbar the effective ion-extraction length scale in the transverse direction is calculated to be approximately 4.5 nm in unit density dimensions. Note that this length scale is comparable with the diameter of a double helix strand, which is 2.3 nm.
2.7.2 Proportional counters

Similarly, proportional counters are also gas detectors just like ionisation chambers. However, proportional counters are designed to multiply the charges from original ion pair productions before detection. The extra step in amplification of the charges permits this type of detector to be used in interactions of low ion pair productions, such as for low energy incident x-ray photons. The ion multiplication process is induced by applications of sufficiently high electric field. There is a threshold of potential difference for the electric field from which the accelerated ions can gain enough kinetic energy to produce further secondary ionisation interactions with bound electrons. Under atmospheric pressure, the value of potential required is of the order of $10^6$ V/m. Thus, any free electrons or charged particles produced in the volume will be accelerated by the field, which will in turn produce more ion pairs upon collisions.

StarTrack

At National Laboratory in Legnaro, Italy [33], another kind of nanodosimeter has been developed. Instead of having the nanodosimetric detector to count ions, as was demonstrated at Loma Linda University and collaborators, they have built an electron counter which is designed to measure the ionisation cluster size distribution as a function of the perpendicular distance away from the primary particle path. The ionisation cluster profiles of alpha particles of energy 5.4 MeV have been resolved from 20 nm to 24 nm in unit-density equivalent dimensions. Recent measurements on 20 MeV proton beams have also been demonstrated.

Jet counter

The Soltan Institute for Nuclear Studies in Poland has developed another variation on the gas-filled interaction chamber as seen in the previous two designs, the Jet counter. The Jet counter, as indicated by its name, is a detector designed to collect ions from pulses of gas injects [6]. It
has been used in measurements to investigate the ion cluster size distribution of alpha particles, in propane and nitrogen gas chambers [122]. The energy range of investigated ionising sources are relatively low, typically from 100 eV to 2 KeV. The sensitive cross section area of the detector is calculated at 2.3 nm x 2.3 nm with 60% collection efficiency.

Overall, all the ion or electron counter designs are successful at simulating the ion-matter interactions at nanoscale. However, they all suffer from the same problem associated with data uncertainties, both caused extrinsically from losses of particles, or internally due to the dissipative nature of resistive electrical components. The design of using many transducers at the detectors inevitably introduces signal losses at each step of the energy transfer. In contrast, the proposed dosimeter design for this project overcomes these limitations and is described in Chapter 5. The following chapter focuses on a series of cryogenic detectors, which are fabricated from low $T_c$ (transition temperature) superconducting materials. Due to the very low operating temperatures, typically below 8K, the noise introduced due to thermal fluctuations are minimised in these detectors.

2.7.3 Solid state dosimetry

The most common dose counter used in radiation protection - thermoluminescence detector (TLD) [70] is an example of a solid state dosimeter. Solid state detectors are designed to capture the damage caused by ionising radiations by recording it as a physical change in the detector material. Radiochromic film is an example of a type of material that react to ionising radiations by formation of colour centers [116]. TLDs consists of a radio-sensitive material made from a crystal such as LiF, CaF$_2$ or Al$_2$O$_3$ and rare-earth elements or transition metals for activation. Upon exposure to incident ionising radiation, ion pairs produced in the material is trapped within the material. The information about the creation of the ion-pair can only be uncovered after heating. This process allows the electrons and holes to recombine and light is emitted as
a result. The emitted light is then detected by multiplier tubes. TLDs are able to detect doses from $10^{-5}$ Gy to $10^2$ Gy and can be reused, i.e. to reset traps, upon being heated.

However, despite having good sensitivity solid state detectors tend to have low spatial resolution. For TLD the spatial resolution is in the order of a few millimetre. They also often lack the ability to measure track structures in real-time.

Micrometer sized solid state detectors are typically made from silicon or CVD diamond [7]. Although germanium or GaAs are also possible but their high Z number (> 30) as compared to the Z of carbon/nitrogen/oxygen-based tissue makes them not so ideal as biologically-equivalent materials ($Z=6 \approx 8$). Due to the small band gaps in the sensitive material used in the detector, they are 10 times more sensitive to smaller energy depositions compared to gas-based detectors. Both silicon-based and CVD-based [18, 100, 146] detectors work on fairly similar operation principles. For silicon-based detectors it is essentially a PIN (Positive-Intrinsic-Negative) diode, which is biased at appropriate potential. Incoming ionising particles interact with the detector by inducing electron-hole pairs. This process is more likely to happen in the intrinsic part than the doped part due to the extra volume the intrinsic part is exposed to. The ionised electrons and holes can be removed from the detector and collected using a drift potential, and the current is easily detected using standard techniques. CVD based detectors work also like an ion-pair collection device as with the silicon-based ones. The radio-sensitive material is a thin sheet of diamond and after exposure the generated charges are tapped off from two metal electrodes that are positioned on its either side. Diamond based detectors are radiation-durable and are closely related to tissues in term of Z-values. Thus, it makes them a good candidate as detector materials.

Nevertheless, these currently discussed solid state detectors are still not suitable for nanodosimetry due to their micron-dimensions. These micro-sized solid state detectors also suffer from degradation with time [146]. The need for a dosimeter of nanometric resolution and ex-
cellent temporal response will be the ideal detector. More information on semiconductor-based
dosimeters can be found in the literature [7, 146].

2.7.4 Cryogenic bolometry

Bolometers are devices that are designed to measure the temperature change in a material due
to some external sources. Regardless of the spatial dimension of the system of interest, all
bolometers work on the same simple principles. Three common components are found in all
bolometers:

1. An energy absorber. Ideally a material that can interact with the incident particle so that
   the incident particle stops within the target absorber.

2. A thermometer. This is the temperature transducer that converts the temperature of the
   absorber (1) into a readable output.

3. A heat bath. This is the key component to maintain the absorber back to the operational
temperature after absorptions of incident particles. The thermal link between the heat
bath (sink) and the absorber determines the time resolution of the device. High conduc-
tance in the thermal link increases the rate of energy transfer and thus reduces the amount
of ‘dead time’ of the device. However, if the conductance is too high then depending on
the detector’s response time the absorption event may not be detectable. Therefore, an
optimisation in the thermal link is required for fast but feasible measurements. At ther-
mal equilibrium, the external power bias to the detector (i.e. from an external heater
from the sampling stage) is balanced by the cooling power of the heat sink (i.e. from the
cryostat).

It can be seen from above that the key in making a sensitive and conceivable detector de-
pends on how optimised the coupling is between a sensitive amplifier and a temperature trans-
ducer. A temperature transducer is a device that turns heat energy into another form of energy. In order to optimise detection and amplification of an incoming signal, impedance matching is a requirement that needs to be considered, especially for cryogenic detectors due to their low impedances.

Superconducting QUantum Interference Device (SQUID) (see chapter 3.5 for more details) are an extremely sensitive magnetic flux to voltage transducer, theoretically reaching quantum limit sensitivity [87, 128, 141], and is thus the most commonly used amplifier used to overcome the low-impedance complications.

## 2.8 Superconductors as calorimeters

Why superconductors?

- Narrow temperature transition range due to superconducting phase transition.
- Can be used as an extremely sensitive thermometer.
- Are commonly used in calorimetry already, such as STJs, KIDs and TESs.

Superconductors undergoes a phase transition from normal, resistive state to the superconducting state at a very specific threshold in temperature, called the transition temperature, $T_C$. The transition window is of the order of $\approx 1$ mK. Figure 2.2 is a cartoon representation of the resistance-temperature characteristic of a superconductor at the superconducting transition temperature. It can be seen that temperature change at around the transition temperature results in a huge change in resistance, and thus in bolometric applications, when the superconductor is biased at its $T_C$ the material can be used as a sensitive thermometer. Further interactions between the superconductor and incident particles, such as scattering of a Cooper electron or quasi-particle, or fluctuations in surrounding system, create disturbances in the
Figure 2.2: The red dot in both figures indicates two different stages in superconducting-to-normal transition. The blue line is drawn to indicates the resistance of a superconductor as a function of temperature. Figure (a) shows the detector just entering the transition where a fraction of the Cooper pairs are exited to the normal state. In Fig. (b) most of the Cooper pairs have reached the normal state.

Superconductivity of the material. Perturbations in the system will break up Cooper pairs in the superconductor, which is the fundamental charge carrier of supercurrent, and reverts the material back towards its normal state. Incident particle, such as scattering of a particle or quasiparticle with the superconducting-material, as shown in Figure 2.2 (b). The threshold to excite a quasiparticle out of its Cooper bound condensate, or in other words, the energy gap of superconductors are typically very small at on the order of $\approx \text{meV}$. The final output of energy absorbed in the detector from the superconductor may be amplified of any type of signals from changing in resistance, impedance, current, power or inductance depending on the detector configuration.

The responsive nature of superconductors to small temperature fluctuation makes them good candidates for applications in particle detection and bolometry in the past 20 years. Advances in cryogenic techniques, such as cryogen-free coolers, in recent years has also facilitated

37
easier and more practical approaches to use these low-temperature detectors. More details on cryogenic detectors can be found in the following chapter.

2.9 Concluding remarks

Gas-based nanodosimetry is a well-known technique to overcome device fabrication difficulties in making nano-sized and reusable radiation detectors. However, the temporal and spatial and energy resolution specifications of the detector can be improved by using other techniques such as cryogenic detectors. We have seen that superconductor-based detectors provides an unparalleled sensitivity to energy perturbations. Due to easy implementations of cooling systems cryogenic detectors may be the next generation of ultra-fast and ultra-resolving energy detectors.

Superconductor-based detectors also excels other dosimeters in terms of detection timing. For the application in nanodosimetry, time resolution to the micro-seconds are desirable and is achievable with superconducting devices.
Chapter 3

Superconductivity basics

This chapter is intended to be a quick guide to the field of superconductivity, fuller descriptions can be found in numerous published textbooks [63, 136]. In this chapter, I begin the discussion with a brief introduction on some of the key concepts.

The phenomena of superconductivity occurs in some elemental metals (Fig. 3.1) and alloys, and they exhibit two distinct features as it transitions through its critical temperature, $T_C$:

- The resistance of the superconductor drops rapidly to zero.

- A perfect superconductor has zero magnetic inductance.

These do not depend on the ways it has reached superconductivity. For instance, in the two scenarios where, firstly, the superconductor is cooled from a temperature higher than its $T_C$ at zero magnetic field, and when the temperature is lower than its $T_C$ the external field is turned, or secondly, with a constant applied field through its transition, it always acts as a perfect diamagnetic, i.e. generating supercurrent within the surface of the material to repel the externally applied field. This is known as the Meissner effect. For the interest of this report I describe the basic concepts in superconductivity, in particular the phenomenological models such as H
Figure 3.1: A table showing the elements in the periodic table that exhibits behaviours of superconductivity under various pressure conditions. Figure reproduced from [31].

The London model and Gorter and Casimir’s two fluid model. This provides sufficient background to understand the operational principles of the cryogenic devices. Detailed discussions are found in numerous superconductivity text books as mentioned earlier.

Phase changes in materials are a familiar concept which are found in every day lives, such as when water turns into steam from a mug of hot chocolate, or from the condensation of water in our freezers. It can simply be thought of as a change of order parameter, from a disordered (high temperature) phase to a more ordered phase (low temperature). The same concept can be translated to superconductivity in the phenomenological models, where the transition happens for the free moving electrons in a metal as it goes from a quasi-particle to paired-up Bose-Einstein condensate. The pairs of electrons are known as Cooper pairs. All the
Cooper pairs form a complicated many-body system, however interestingly, all the Cooper pairs can be described by just one wavefunction (Eq. 3.1). This is just one complex number, $\Psi$, where the amplitude is the square root of the density of those Cooper pairs ($n_s$), and its phase $\phi$.

$$\Psi = n_s e^{i\phi} \quad (3.1)$$

Two fundamental length scales are used to describe superconductors:

1. London penetration depth, $\lambda$
2. Pippard coherence length, $\zeta$

The two stated length scales above are the keys to understand superconducting devices, in particular it is useful in the further discussions on our ISTED device. We now describe a brief derivation and the physical meanings of the two parameters. In the model of F & H London a superconductor consists of normal electrons and superconducting electrons, at temperatures below its $T_C$. This can be considered as a mix of normal fluid and superfluid components, known as the two fluid model. Denoting the number density of normal electrons and superconducting electrons as $n_s$ and $n_{ns}$, and each having velocities $v_s$ and $v_{ns}$ respectively, we can write the motion of a superelectron being accelerated by an electric field, $E$, as follows:

$$\frac{m}{\partial t} \partial v_s = -eE \quad (3.2)$$

$$J_s = -en_s v \quad (3.3)$$

Substituting Eq. 3.3 into Eq. 3.2 we would arrive at the rate change of supercurrent from the superfluid:

$$\frac{\partial J_s}{\partial t} = \frac{n_se^2}{m} E \quad (3.4)$$
One of Maxwell’s equation for electromagnetism states that a changing magnetic field in a coil induces an emf such that the generated current opposes the changing field, also known as Faraday’s law:

$$\nabla \times E = -\frac{\partial B}{\partial t}$$

(3.5)

In light of the Faraday’s equation the dynamical equation for the supercurrent, Eq. 3.4, becomes:

$$\frac{\partial}{\partial t} \left( \frac{m}{n_s e^2} \nabla \times J_s + B \right) = 0$$

(3.6)

It can be deduced that the time dependent function on the left hand side (LHS) of Eq. 3.6 is a constant. In the F & H London model the constant is postulated as zero.

$$\frac{m}{n_s e^2} \nabla \times J_s + B = 0$$

(3.7)

Eq. 3.7 can be rewritten just as a function of the magnetic field, $B$, using another Maxwell’s equation \(^1\), $\nabla \times B = \mu_0 J_s$, and invoking Stoke’s theorem. The Maxwell equation mentioned is also known as Ampere’s law. This states that the magnetic field when integrated around a closed loop of wire creates a current within the wire.

$$\nabla^2 B - \frac{\mu_0 e^2}{m} B = 0$$

(3.8)

It can be seen clearly that Eq. 3.8 is just a second order homogeneous linear equation. The general solution, $B(x)$, to such equations is simply of an exponential form as in Eq. 3.9, where $B_0$ is the magnetic field at the superconductor interface (when $x = 0$) and the spatial variable $x$ increases from 0 (at the interface) in the direction perpendicular to the plane of the superconductor.

\(^1\)\text{$\mu_0$ denotes the vacuum permeability.}
\[
\mathbf{B}(x) = B_0 e^{-\frac{x}{\lambda_L}}
\]

(3.9)

where,

\[
\lambda_L = \sqrt{\frac{m}{\mu_n e^2}}
\]

(3.10)

Eq. 3.9 suggests that the rate of spatial decays of the magnetic field into a superconductor depends only on the value of \(\lambda_L\) in the exponential form. This value is the London penetration depth, defined as the perpendicular length needed for the magnetic field to decay to \(\frac{1}{e}\) th of its initial value at the interface. \(\lambda_L\) is a function of the number density of super electrons, and since the ratio of normal to super electrons is determined by the temperature the number densities is also a function of temperature. This is characterised by the Gorter and Casimir two fluid model as shown in Eq.3.11.

\[
n_s \propto \left[1 - \left(\frac{T}{T_C}\right)^4\right]
\]

(3.11)

Now we can finally write \(\lambda_L\) as a function of temperature, where we use \(\lambda_0\) to denote the penetration depth at zero temperature.

\[
\lambda_L(T) = \lambda_L(0) \frac{1}{\sqrt{1 - \left(\frac{T}{T_C}\right)^4}}
\]

(3.12)

Whilst the London penetration depth describes the spatial rate of decay of a magnetic field inside a superconducting material the coherence length, \(\zeta\), describes the characteristic decay length (the exponent) of the Cooper pair wavefunction into a different medium. This is the characteristic length scale over which a supercurrent is maintained outside the superconductor. \(\zeta\) is determined by the energy gap between the condensed and excited states of the Cooper pairs, \(E_g\), and its Fermi velocity, \(v_F\). Equation 3.13 shows the coherence length, \(\zeta_0\), in zero field condition.
Figure 3.2: Illustrations on how the field penetration amplitude decays for type I (a) and type II (b) superconductors.

\[ \zeta_0 = \frac{\hbar v_F}{\pi E_g} \]  \hspace{1cm} (3.13)

With the two characteristic length scales established it is now a good moment to discuss the general categorisations of superconductors: type I and II. Type I describes the case when \( \zeta \gg \lambda \), meaning that the superconductor screens all the external fields as the penetration depth is much smaller than the coherence length. This is a non-local electrodynamical behaviour and is typically applied to pure metals. Under a given flow of supercurrent the applied field, \( H \), required to break the supercurrent in temperatures lower than \( T_C \) is illustrated in Fig. 3.2. In contrast, type II superconductors have a \( \lambda \) much greater than \( \zeta \), typically found in impure metals or alloys. At field strength less than \( H_{c1} \) complete field expulsion is achieved. Between \( H_{c1} \) and \( H_{c2} \) penetration of field lines are allowed in the form of quantised vortices, known as fluxoids. One fluxoid, \( \Phi_0 \), is a fundamental parameter of value \( \frac{\pi \hbar}{e} \).

### 3.1 Josephson Effect

Josephson junctions are important building blocks for all quantum devices including SQUIDs, therefore it is a natural roadmap for us to divert into the basic principles of a Josephson junction.
Josephson effect refers to the continuous flow of supercurrent between two distinct and separated superconductors. The gap in between the two superconductors can be anything; such as metals, semi-conductors, dielectrics (insulators), air, vacuum, or even grain boundaries. In general a junction is anything that causes a change in current density of the electrons in the superconducting state. When two superconductors are in close proximity to one another a supercurrent is allowed to pass. This is a remarkable phenomena. If we take a piece of copper and observe its resistance as we cool down its temperature, we find that there will always be a finite resistance across the metal even at very low temperatures. However, counter-intuitively when you insert a piece of copper in between a small gap of two superconductors then somehow a supercurrent is allowed to flow, with no dissipation. This behaviour is very similar to the tunnelling of a normal electron in a simple metal system, where the wavefunction of the normal electron overlaps over a tunnelling barrier and thus allows the electron to transverse. By analogy with the simple electron system the phenomenon of supercurrent through a weak link can also be thought of as the overlaps of wavefunction of the Cooper pairs. Figure 3.3 illustrates the distribution of the amplitude squared of the Cooper pair wavefunctions on both sides of the gap. The wavefunction amplitude is flat on both superconductors as it should be a constant representing the order parameter of the material. As an electron from the Cooper pairs enter the superconducting-metal (S-M) interface, for an evanescence amount of time the Cooper pairs remains coupled to each others’ phases and it is assumed that its wavefunction modulus decays exponentially inside the gap. The rate of decay is determined by the properties at the interface. However, as long as the weak spot, the lowest point on the sum of the wavefunction amplitude squared from the left and right sides of the gap (shown in Fig. 3.3 as the dotted line) does not reach zero supercurrent is allowed to transmit across.

The phase difference of the Cooper pair, $\phi$, across the junction is the most important parameter that determines all the properties of a Josephson junction. This is defined as simply the
Figure 3.3: A schematic diagram of the distribution of the square of the wavefunction amplitude (the number density) of Cooper pairs at a Superconductor-Metal-Superconductor (SMS) junction.

phase on the left hand side superconductor, $\phi_1$, minus the phase on the right hand side, $\phi_2$, as shown in Eq. 3.14.

$$\varphi = \phi_1 - \phi_2$$  \hspace{1cm} (3.14)

$\varphi$ is used in describing the current allowed to pass through a JJ. This current-phase difference relation under zero field is of the form in Eq. 3.15, where $I_C$ is the maximum allowed supercurrent:

$$I = I_C \sin \varphi$$  \hspace{1cm} (3.15)

This relation is simply a sinusoidal function. The magnitude of the current is at a maximum when the phase difference is of integer increment of $\pi$ from $\frac{\pi}{2}$, and minimum when $\varphi$ is at multiple integers of $\pi$. In other words, Eq. 3.15 suggests that when the Cooper pairs across the
juncture are either in phase or are the same the supercurrent is broken. This equation is known as
the DC (direct current) Josephson effect as a DC current can be realised in a JJ, in the absence of external electric or magnetic fields.

Another key equation, describing a JJ is known as the AC (alternating current) Josephson effect, see Eq. 3.16. This describes how an AC current can be realised in such junctions by applications of a DC voltage bias across the junction. This is a useful relation which is applied to metrology to define the fundamental value of $\frac{h}{e}$, or as a frequency to voltage transducer.

$$2eV = h \frac{\partial (\Delta \phi)}{\partial t}$$ (3.16)

### 3.2 Current-Voltage (IV) characteristic

The electronic characteristics of a Josephson junction is discussed in this section. As we will see shortly, a JJ has its own unique IV characteristic just like any other electronic components, such as diodes, inductors or capacitors. A JJ can be incorporated into useful circuits with appropriate logic rules if we know its current-voltage characteristics.
From the DC Josephson equations (Eq. 3.15) we see that when the current is below $I_C$, $\Delta\phi$ is constant. When we feed this information into the AC Josephson equation (Eq. 3.16) we see that since $\frac{\partial(\Delta\phi)}{\partial t} = 0$ the voltage across the junction is also zero, i.e. $V = 0$. In contrast, when $I \gg I_C$ phase becomes time dependent and thus, $\frac{\partial\Delta\phi}{\partial t} \neq 0$ and so $V \neq 0$. This can be seen as a delta step in voltage at $I = I_C$, shown in Fig. 3.5. At currents greater than $I = I_C$ the Cooper pairs behave in its normal states and thus the IV resembles one of a normal resistor with the IV line extrapolating to the origin. However the same IV characteristic is not reproduced as the current is swept back to zero. This is known as a hysteretic effect. When the current is reduced back to $I_C$ there exists a momentum from the resistive current such that the Cooper pairs are not in its condensate until the current reaches the retrapping current, $I_r$. Arrows in Fig. 3.5 indicates the direction of the curve as current is swept up and then down in the first quadrant. The hysteretic effect creates a problem if such devices were used in a circuit. However it can be eliminated by introducing a shunt resistor across the junction, which we shall see in the next section.
3.3 Resistively Capacitively Shunted Junction (RCSJ) model

The RCSJ model is often used to conceptualise the electrical properties of a real JJ. In this model, the junction is seen as a composite of a JJ, resistor and a capacitor connected in parallel as shown in Fig. 3.6, where the cross represents a JJ. In the discussion in the previous section it is now clear to see why a good quality JJ shows prominent hysteresis. In practice, the resistor and the capacitor reflect the junction shunting resistance and the geometric shunting capacitance in a JJ respectively. In practice, the unwanted hysteresis in a JJ is accounted for by introducing a shunt resistor as a dissipation component. This allows resistive dissipation in the circuit and not affecting the transition current of the JJ component.

The circuit shown in Fig. 3.6 is very much like an RC (resistor-capacitor) circuit except for an additional JJ. In principle, one could work out the characteristic time constant or frequency for such a circuit. This term is, in fact, an important parameter to describe the level of hysteresis of a real JJ junction. This is called the Stewart & McCumber parameter, $\beta_C$. I will revisit this concept again in Chapter 8 when I examine the intrinsic noise in a nanoSQUID. Below I present the derivation of the time-dependence of phase in presence of an applied current, before arriving at an expression for $\beta_C$. 
By Kirchhoff’s law the total current through the junction is as follows, where the current through the JJ is taken from the DC Josephson equation (Eq. 3.15):

\[ I = I_C \sin \phi + \frac{V}{R} + C \frac{dV}{dt} \]  

(3.17)

With the aim of arriving at an equation in terms of \( \phi \) in mind, we can eliminate the voltage dependence in Eq. 3.17 by a substitution of the AC Josephson equation (Eq. 3.16), and rearranging as follows:

\[ I - I_0 \sin \phi = \hbar \frac{1}{2eR} \frac{d\phi}{dt} + \hbar \frac{C}{2e} \frac{d^2\phi}{dt^2} \]  

(3.18)

The left hand side of the above equation can be written as a first order differential equation of an arbitrary function \( U \) with partial respect to \( \phi \), as follows:

\[ I - I_0 \sin \phi = -\frac{2e}{\hbar} \frac{\partial U}{\partial \phi} \]  

(3.19)

where \( U = -\frac{\Phi_0}{2\pi} (I\phi + I_0 \cos \phi) \).

The \( \phi \) dependent function \( U \) in Eq. 3.19 can be seen as the dynamics of a ball rolling on a ‘stepped downhill’ potential function, or it is more commonly being referred to as the ‘tilted washboard’ potential (Fig. 3.7). When the applied current is less than \( I_C \) local minima can be seen in the potential function, creating conditions for the particle to be confined to one of the ‘wells’. This is known as the static case. The particle oscillates in the well with frequency, \( \omega \):

\[ \omega = \omega_0 \left(1 - \left(\frac{I}{I_0}\right)^2\right)^{\frac{3}{4}} \]  

(3.20)

where \( \omega_0 \) is a constant defined as

\[ \omega_0 = \left(\frac{2\pi I_0}{\Phi_0 C}\right)^{1/2} \]  

(3.21)
Figure 3.7: The tilted washboard potential for $I < I_C$ (dotted) and $I > I_C$ (solid) for a ball traveling in the phase space.

With analogy with the particle oscillating in simple harmonic motion inside the well, the average of its displacement, i.e. its phase, is a constant. This means that the rate change of phase is zero, suggesting that voltage is zero too according to the AC Josephson equation (Eq. 3.15). As a sanity check, we are considering the case when $I < I_C$ thus we should expect a zero voltage drop since Cooper pairs are in the condensate state.

Now in the case when $I > I_C$, the potential function in the phase space in Eq. 3.19 changes to a form as shown in Fig. 3.7 as the solid line. In this case, the local minima of the potential flattens so that the particle rolls down the hill. In other words, the phase of Cooper pairs is switching from the static case to have a time dependent term. Since the rate of change phase is proportional to voltage we also see a developing voltage across the junction. Imagine now that we reduce the applied current. The reverse effect happens where the wells are starting to develop. In an idealised junction with no hysteresis the particle becomes trapped instantly into the nearest well. In a not-so-desired junction the particle would continue rolling until the dissipative energies are greater than its initial inertia. At this point the particle confines to a well and the current applied is called the retrapping current, $I_r$. The dissipative forces in
the particle-washboard analogy is analogous to the damping term of the RCSJ system. This parameter is named by Stewart and McCumber as $\beta_C$ (Eq. 3.22).

$$\beta_C = \frac{2\pi R^2 C}{\Phi_0}$$  \hspace{1cm} (3.22)

It is a natural tendency for any physicist to examine the limiting cases of the characteristic parameter of a system. Hence in this case, the two limiting cases are as follows:

- $\beta_C \ll 1$. The strongly overdamped limit.
- $\beta_C > 1$. The strongly underdamped limit.

Without going into the mathematical treatments of the indicated conditions one finds that in the strongly over-damped case, the averaged current and voltage follows for both the forward and backward sweeps coincides completely, i.e. the re-trapping current is $I_C$. However, in the other scenario the IV characteristic has varying degree of hysteresis depending on the value of $\beta_C$. The greater the value of $\beta_C$ the more pronounced the hysteresis, i.e. the greater the re-trapping current.

### 3.4 DC-SQUID and flux quantization

In the presence of an external magnetic field, as the temperature drops below $T_c$ of a superconductor loop the magnetic field in the loop remains there even when the external field is switched off. This is due to the persistent supercurrent generated in the loop and it only generates magnetic field of integer multiples of a constant, flux quantum. Quantisation of magnetic flux was first predicted by London.
3.5 Superconducting QUantum Interference Device (SQUID)

Building on top of the components in superconductivity discussed earlier, a SQUID consists of a loop of superconducting wire with two Josephson junctions in parallel, as shown in Fig. 3.8. The concept of developing particle detectors with superconducting material is a rapidly growing field, driven by the needs for applications such as in quantum information and single photon metrology [61]. SQUIDs are commonly used in cryogenic devices as a read out component due to its low impedance. In this section I will use the relevant physics concepts discussed in previous sections to explain the operation principle of a SQUID.

The SQUID consists of a superconducting loop that, in simple electronic terms, acts as a current divider. Each branch receives a portion of the current, $I_B$, with respect to the ratio of impedances of each branch. In the most ideal operation condition both branches are fabricated to have the same impedance, thus divides $I_B$ equally. The critical current of this parallel Josephson junction (JJ) loop is therefore twice the value of a single JJ. In the case where there is no applied magnetic field, and with $I_B < I_C$ (from now on, I will use the term $I_C$ to represent
the critical current of the SQUID loop rather than a single JJ), the supercurrent is maintained as the order parameter through both JJs are unchanged.

However, when the SQUID is placed in a magnetic field perpendicular to the planes of its loop, and with the same condition that \( I_B < I_C \), we see that the SQUID loop responds to the external field by generating an additional current to oppose the field. This current is known as the screening current, \( I_{\text{screen}} \). Now it is clear to see that the current through the two branches are no longer equal - a break in the SQUID’s symmetry. In the specific case illustrated in Fig. 3.8 the branch on the left and right, \( I_{\text{left}} \) (Eq. 3.23) and \( I_{\text{right}} \) (Eq. 3.24) respectively, would differ by \( 2 \times I_{\text{screen}} \). The current through the JJ on the left is now greater than the one on the right.

\[
I_{\text{left}} = \frac{1}{2} I_B + I_{\text{screen}} \quad (3.23)
\]
\[
I_{\text{right}} = \frac{1}{2} I_B - I_{\text{screen}} \quad (3.24)
\]

By ramping up the magnetic fields \( I_{\text{screen}} \) increases in response. This relationship breaks until one of the JJ (left in this case) is driven to the normal state. All the current now only passes through the other JJ since it is the one with the least (zero) resistance, until the current also surpasses the second JJ’s critical current. It is at this point something interesting happens. If the field is continued to ramp up, the SQUID does not become resistive to maintain a complete exclusion of magnetic flux through its loop, instead one flux quantum, \( \Phi_0 = \frac{\hbar}{2e} \), amount of flux is allowed to penetrate into the loop. The SQUID effectively resets itself so that the process of generating \( I_{\text{screen}} \) is repeated until both JJs reach their junction critical currents again. The SQUID’s screening current is periodic with one period equal to one flux quantum.

The SQUID can be used accurately as a flux to voltage transducer due to its large range of linear voltage response to magnetic fields. In order to operate the SQUID in its most sensitive region one needs to optimise the SQUID’s bias current. This is the region of the steepest voltage
response to changes in field. This typically happens when the voltage response over a period of $\Phi_0$ is the largest, at a current just above its zero field $I_C$.

The electrical properties of a SQUID has been exploited for the use in electronics as a sensitive flux-to-voltage transducer. This is achieved by connecting many elements of SQUIDs in series, forming a SQUID Series Array (SSA). The advantage of SSA over a single SQUID is seen through its significant higher gain and larger operation bandwidth [144, 34]. It also benefits from the increase in its impedance, which makes electronic implementation easier. In chapter 8, I describe the use of an SSA made from a 100 series arrays of DC SQUIDs for a measurement on the intrinsic noise limit of a nanoSQUID device.

3.5.1 Noise in DC SQUID

The sensitivity of a detector depends on its susceptibility to noise. The more robust it is against pick-up noise the more precise and accurate the detector can measure. In the following discussions we describe the important sources of noise in a SQUID device. This is useful and motivates for our future discussions in Chapter 8 on the results I have obtained for nanoSQUID noise measurements.

The focus of this section is on the discussion about thermal noise and low frequency noise.

3.5.2 Thermal noise

Noise can originate from the thermal fluctuations due to a flow of current in any resistive components. In the case with a SQUID it is the presence of the shunt resistance in the RCSJ model. This type of noise is known as the thermal noise, Johnson or Nyquist noise. This is a major noise source contribution for a SQUID device. Thermal fluctuations affects the stability of the electron density, which consequently causes degrading in the voltage read-out precisions. In the junction’s resistive region of operation, $I \gg I_0$, both the current and voltage fluctuations
contributes towards an observable rounding in the junction’s transition at $I_C$.

Voltage fluctuations depend on the temperature and resistance of the electrical component [34], however for an ideal resistor it appears as a constant term in the frequency spectrum. It can be taken as a Gaussian distribution of appropriately scaled random numbers [34]. Equation 3.25 describes the voltage noise spectral density, $S_V$, where $R$ is the resistive term from the shunting resistance of the junction, $T$ and $k_B$ has the usual meaning for temperature and Boltzmann constant. The unit-less component associated with the down-mixing of Johnson noise is accounted for by the constant $\gamma$, typically of value 8.

$$S_V^{1/2} = \gamma k_B T R \quad (3.25)$$

From the voltage noise spectral density in Eq. 3.25 we can convert it to the flux noise spectral (Eq. 3.26) density through a conversion factor, $V_\Phi$. $V_\Phi$ is the SQUID’s voltage response to a change in flux of one flux quantum ($\Phi_0$).

$$S_\Phi(f) = \frac{S_V(f)}{V_\Phi^2} \quad (3.26)$$

A useful technique to compare different SQUIDs’ performances is to standardise $S_\Phi(f)$ in terms of energy noise spectral density, since $\Phi_0$ is dependent on the geometry of individual SQUIDs.

$$\varepsilon(f) = \frac{S_\Phi(f)}{2L} \quad (3.27)$$

With respect to the washboard potential model discussed in an earlier section the current fluctuations can be seen as a fluctuation in the tilt of the washboard. At the system’s sensitive region when $I$ is approaching $I_C$ the additional noise current may excite the particle into another local minimum potential, especially in the case when the damping term, $\beta_C$, is much less than 1.
This is observed as a series of randomly distributed voltage jumps in time. The time averaged voltage with applied current just less than the junction $I_C$, i.e. when $I < I_C$, is measured as a finite value and hence we obtain a rounded IV feature at the transition current. Moreover, for a junction with a finite shunting capacitance the undesired effect due to thermal fluctuations in effect also suppresses the hysteresis effect.

To quantify the contributions of the thermal fluctuations to the overall noise one often use the unit-less parameter, $\Gamma$, to represent the ratio of thermal energy against the Josephson coupling energy.

In the case when $\Gamma$ is much less than 1, i.e. $\Gamma \ll 1$ thermal fluctuations does not play an important role in the noise contribution, and when $\Gamma$ approaches one we are in a regime of strongly decoupled Josephson effects.

**Flicker noise**

Noise at the low frequency operational range of a SQUID is dominated by the $1/f$ or flicker noise. This is particularly significant for low frequency SQUID applications such as biomagnetism and geophysics as the frequency range of interest are at 0.1 Hz or below.

For an superconductor-insulator-superconductor (SIS) junction the sources of $1/f$ noise degrades the sensitivity of the junction through trapping of electrons in defects at the tunnel junction [86], or at the superconducting material [86, 87], or sometimes the trapping of flux lines in the SQUID body [62].

### 3.6 Cryogenic detectors

As opposed to room temperature solid-state detectors, cryogenic detectors are significantly more sensitive due to reductions in the thermal noise at the lowered operating temperature
range. At operating temperatures that are approaching the absolute zero much more sophisticated apparatus are required to reach and maintain such low temperatures. With the advances in cryogenic systems, such as cryogen-free pulse-tube cooling systems that are capable of reaching sub-100 mK in temperature, it is becoming easier and more feasible to implement low temperature experiments. Cryogenic systems are further discussed in Chapter 7.

In the following sections, I will discuss the basic design concepts behind some cryogenic detectors that are capable of bolometric measurements, namely Transition Edge Sensor (TES) and Kinetic Inductance Detector (KID).

3.6.1 Transition Edge Sensor (TES)

Within the superconductive detector community the Transition Edge Sensor (TES) is one of the mostly researched devices. As the name suggests, TES consist of superconducting materials that are operated close to its transition points. They are typically current biased and are used in two ways: either they are directed configured as a particle detector, or indirectly designed to detect particles through phonon mediations. In the case of direct measurement, it behaves in a similar way to Superconducting Tunnel Junctions (STJs), where the incident particle interacts with a superconductor directly and breaks Cooper pairs in the process. This perturbation can be measured as a change in the resistance of the superconductor. In the case of indirect detection, an incoming particle interacts with the lattice of an absorber instead. The interactions vibrationally excites the absorber lattice, which results in productions of phonons. Perturbations in the absorber’s phonon system is coupled with the phonon system of the superconducting film, which is consequently linked to the superconducting film’s electron system and to the detectable output signal. An absorber is used as the interacting medium and it is especially useful when the stopping power or the heat capacity of the superconducting film is not enough to detect incident particle in the desired energy range.
Given the versatile uses of superconducting materials in sensing, each TES is designed specifically for its application. TES is used extensively in the astrophysics community for X-ray photons detections as well as the detection of cosmic microwave background polarisations. The device is typically read out by Superconducting QUatum Interference Devices (SQUID) as discussed in section 3.5.

A typical TES calorimeter works in the energy range of roughly 100 KeV or less, with good resolution of 10 eV but very long dead time of 500 $\mu$s. The heat absorber in a TES consists of a material that varies in resistance with different ambient temperatures. It is maintained at operating temperature by voltage bias the superconductor to achieve negative electrothermal feedback [76]. Since the sensor is supplied with an electric current the detector’s sensitivity would suffer from electrically related noise. The ultra-low operation temperature required for this device, of typically 100 mK, makes this device less practical to operate in conjunction with an ion beam source.

Several groups are developing TES for a range of particle detections, however only a few are working in the MeV energy range. Recent activities [29] in MeV detections were conducted jointly by groups at Los Alamos National Laboratory (USA), National Institute for Standards and Technology (NIST, USA) and STAR cryogenics (USA). The group has reported a capability of energy resolution for alpha particle detection at 1.17 +/- 0.08 KeV FWHM at 5.3 MeV. The detector is operated at 120 mK. However the detector is limited by its size of $4 \times 4 \times 0.25$ mm and a total detection time of 24 hours. With those limitations it is not feasible to be used as a nanodosimeter.

3.6.2 Kinetic Inductance Detector (KID)

Similarly to TES, Kinetic Inductive Detector (KID) is also a type of photon detector employed mostly in astrophysics [30]. It can be used to resolve the energy and arrival time of incident
particles, furthermore, the main difference in KID as compared with other low temperature calorimeters is that it can be easily multiplexed into large arrays.

KID works by using the dependence of the kinetic inductance of a superconductor as a function of the density of Cooper pairs. A meandering superconductive strip is typically used as the sensitive absorber of the detector, and is part of a resonant circuit configuration. Absorption of an incident photon causes some Cooper pairs to break which, as a consequence, produce a number of quasiparticle excitations. The change in the Cooper pair density then causes a change in the kinetic inductance of the strip, creating super currents in the superconductor. This super current can be thought of as stored information, and can be read by knowing the current required to reverse it, accounting for the extra inductance the reversed current introduces. This read out system is usually a capacitor, made from a thin film superconducting resonant system, forming a microwave resonator that has a variable resonant frequency depending on the energy absorption of the photon.

It was first developed at the California Institute of Technology and the Jet Propulsion Laboratory in 2003 for the purpose of photon detections, in the frequency range of the far-infrared to X-rays. However, again this type of detector operates at ultra-low temperatures of 100 mK. The optimal energy detection is far below the energy used in medical radiotherapy.
Figure 3.9: Schematic diagram of a KID element.
Chapter 4

Nanodosimetric quantities on sub-1keV electrons

1 Low mass ionising particles of low energy levels are considered to be key contributors towards the causes of radiobiological damages at present, and thus the interactions of these particular low energy ionising particles are important in nanodosimetric measurements [69]. It is thought that the rate of cell death is related to the number of lesions that took place within the dimensions of a DNA segment. The more lesions there are the more complex the damage sites will be, and consequently the harder it is for the cell to self-repair. This means that ionising particles that cause more severe biological damages are the ones with mean free path that is comparable with the length scale of interest for a DNA strand. Mean free path is defined as the

1The simulation and analysis using GEANT4-DNA presented from section 4.3 are solely my contributions. I have built my simulation on top of GEANT4-DNA examples and I am grateful for the contributions from the GEANT4-DNA working group for their work and in making the source codes publically available. In the sections prior to 4.3, I have presented works from the literature on nanodosimetric quantities.
average distance between each interaction for an ionising particle of certain energy. A DNA strand is a double helix structure with a diameter of approximately 2 nm. Whilst the mean free path for high energy ionising particles are orders of magnitude greater than nanometer, the mean free path length for light ions such as Helium, protons and electrons with low energy are more comparable with biological sensitive length scales. For light ions, low energy regime is between 100 keV and 20 MeV [69], and for electrons the low energy regime is for energies below 1 keV. More information on the categorisation of radiobiological damages can be found in section 4.2.2.

Common interactions between low energy light ions and condensed matter are electromagnetic processes such as ionisation and excitation. Elastic interactions, on the other hand, are of lower cross section and results in little scattering. This means that they can often be neglected [91]. Charge-transfer interactions that lead to formation of anions are also neglected due to their low interaction probability [124].

Particle track structure is more conveniently studied from computational simulations. They are frequently used to study low energy ionising particle interactions with condensed matter as they are easier and quicker to implement in comparison with experiments. The calculations from most of these particle track simulations are based from classical concept on particles and particle interactions. However, as classical mechanics breaks down when the energy level of interacting particles becomes small it is essential to examine its validity for the simulation of nanodosimetric particles.

To begin the examination on the validity of classical mechanics on radiobiological relevant particles it is important to introduce a basic concept in quantum mechanics- de Broglie wavelength. All objects, whether macroscopic or microscopic, has an associated de Broglie wavelength, denotes as \( \lambda \). It is a function on the object’s momentum \( p \) and velocity \( v \) in a simple relationship shown in Eq. 4.1,
where $\hbar$ and $m$ represents the Plank’s constant and particle mass respectively.

A quick examination of Eq. 4.1 tells us that the de Broglie wavelength becomes large when the interacting particle has small mass or when the particle is travelling with low velocity. For an example, let’s take a 10 eV electron and a proton of $\approx 20$ keV of kinetic energy. Due to the mass difference between the two particles the velocity for both particles are actually roughly the same. Both have a de Broglie wavelength $\lambda$ of 0.39 nm, which is comparable to the diameter of the target molecule of interest - liquid water. An electron of 30 eV has a de Broglie wavelength comparable to the diameter of Carbon atoms, which is an element that is vital to the formation of biological matter. With an energy of 150 eV for electrons and 300 keV for protons the de Broglie wavelength reduces to $\approx 0.1$ nm, which is the lengthscale for bond length between Carbon atoms [145]. At interactions of even lower energy levels, ionisation interactions becomes less important in relation to electronic, vibrational excitation and charge-transfer interactions. For instance, the dominant interactions for ultra-low energy electrons of several eVs are elastic, rotational excitations and vibrational interactions. Slow projectiles are more likely to transfer a significant amount of momentum to the target molecule, which results in a change of its direction of motion.

The most common approach to compute track structure simulations uses Monte Carlo methods. Monte Carlo methods will be covered in detail in section 4.3.1. Outputs of such simulations includes the location and transfer energy for all interactions provided that the energy transfer is above an user-defined energy cut-off level. The computed interactions covers both elastic and inelastic interactions.

However, there are problems to the validity of track structure simulations as Fano has shown that interactions between low energy projectile and condensed medium doesn’t have
well-defined interaction location. Instead, it makes sense to be thinking about the exact in-
teraction locations in the classical sense as being decolalised [44]. There are two reasons for
his argument. Firstly, acording to quantum mechanics the de Broglie wavelength of low en-
ergy micro-particles becomes comparable with the intermolecular spacing in the interaction
medium which means the particle will start to behave quantum mechanically. Secondly, col-
lective oscillations/excitations that occurs in the target medium [43] after an interaction makes
it irrelevant to pin-point the exact interaction location. For instance, an excitation interaction
that is localised to one of the interacting particles for a time period of $10^{-16}$s [104] can be quickly
delocalised in the subsequent time frame [79]. It is, therefore, not possible to resolve the exact
location of the interaction more precisely than the de Broglie wavelength of the particle, which
is also satisfied by Heisenberg’s uncertainty principle.

Nonetheless, as we shall see in section 4.1.2, Liljequist et al. challenged the hard rule on
the validity of using trajectory simulation on low energy particles. The validity condition as
proposed by Liljequist et al. says that it is not always strictly necessary that the wavelength of
the ionising particle should be much smaller than the intermolecular spacing of the interacting
medium. Instead, the much greater-than relation between the particle’s de Broglie wavelength
and intermolecular spacing is replaced with simply a greater-than relation.

This chapter is organised into the following three sections:

1. Quantum-classical limit.

2. Important nanodosimetric quantities.

3. Effects of spatial delocalisations to ionisation cluster-size distributions.

In the section Quantum-classical limit, I will examine the limitations in classical trajec-
tory simulations and introduce the concept of circumstantial validity of trajectory methods as
suggested by Liljequist et al. In a nutshell, the circumstantial validity imposes a softer rule
on the validity of trajectory methods and permits its use on simulating low energy ionising particles that are of interest in nanodosimetry when conventional validity rules would forbid it. In section **Important nanodosimetric quantities** I will introduce some key nanodosimetric concepts such as biological sensitive volumes, categorisation of ionising radiation induced biological damages and measurable nanodosimetric parameters. These important nanodosimetric concepts will be useful in the discussion in the subsequent section. In the final section of this chapter, **Effects of spatial delocalisations to ionisation cluster-size distributions**, I shall examine the effect of spatial uncertainties from Heisenberg’s uncertainty principle on ionisation cluster-size distributions for sub-1 keV electrons in liquid water. In this section we will also review Monte Carlo methods and the simulation environment and experiment procedures in GEANT4-DNA.

### 4.1 Quantum-classical limit

One of the motivations for studying interactions between low energetic electrons (below 1 keV) and condensed matter is due to their importance towards the understanding of biological outcomes post irradiation events. It has been shown that electrons of a few hundred electron volts are likely to be causing clustered damages to target volumes on the nanometer scale, which coincides with the important biological lengthscale - a DNA strand, and thus making them an important contributor towards DNA damages and cellular outcomes [108, 111, 113]. The study of low energy electrons are also useful in other fields such as in Auger electron spectroscopy, photoelectrons spectroscopy, and secondary electron emission theory.

Many particle track trajectory simulations are computed based on classical mechanics, where particles are treated as classical objects with well-defined and localised position and momentum. However it is well known that classical mechanics breaks down for extreme cases and
should be replaced with more appropriate theories. For instance, the de Broglie wavelength for electrons of energy 100 eV or lower is greater than inter-molecular distances in liquid water. This means that for low energy electrons quantum effects becomes particularly important. Under quantum mechanical considerations, for instance, scattering interactions are interpreted as multi-centred coherent scattering as the incident particle’s wavelength stretches across several target molecules. Therefore, it can be seen that the validity of using the classical treatment for calculating track structures becomes debatable [135].

Common track-structure simulations adopts the implementations of Monte Carlo methods. The target medium is assumed to be spatially homogeneous and continuous. Non-static particles are also assumed to move freely without disturbing the target material until it undergoes an interaction, either elastically or inelastically, with the target. The probability for the particle to traverse without encountering a single interaction decreases as the distance it is moved by increases, as shown in Eq. 4.2.

\[ P(s) = e^{-s/\lambda_t} \]  

(4.2)

where the relation between distance moved, \( s \), and the likelihood of finding the particle \( P(s) \) with a given \( s \) is dependent on the total mean free path, \( \lambda_t \). The total mean free path is defined in Eq. 4.3, where \( \rho_t \) is the total cross section per scatterer object, and \( n \) is the density of the target medium.

\[ \lambda_t = (n\rho_t)^{-1} \]  

(4.3)

Simple trajectory simulation assumes that scattering interactions takes place on a single target atom or molecule. However, for interactions such as plasmon excitation, phonon creation and annihilations where a collective of target objects are of interest, one needs to find alternative methods to simulate these interactions. Possible approaches to determine the cross section
profiles of these interactions can be found through either applying some standard quantum mechanical calculations or by basing the outcome of these interaction on empirical results.

Despite the availability of cross section data for interactions of low energy electrons with liquid water in the GEANT4-DNA package the precision and accuracy for this data is not very good [38, 39, 114, 95]. The uncertainties associated with elastic interactions are particularly great. Therefore, it is clearly seen that the use of trajectory methods are hugely limited by the availability of quality cross section data. It is also limited to simulations of particles within the classical realm. For instance, it cannot simulate an electron diffraction pattern when the energy of the particle gets sufficiently small that the quantum nature of the particle becomes more dominant.

The boundary between quantum and classical mechanics are usually determined by the ratio between the incident particle’s de Broglie wavelength, $\lambda$, and the mean inter-molecular distance within the scatterers, $d_{nn}$, which is approximated as $d_{nn} = n^{-1/3}$. In the case when the simulated particle falls into the quantum regime then a complete quantum mechanical treatment is required for the calculation of the particle’s dynamics [120]. Such methodologies has been applied to simulate an electron’s multiple elastic scattering processes in a target medium of DNA molecules coherent multiple elastic scattering [20, 21]. However, the disadvantages of using quantum mechanical simulations are that it is relatively much more computationally expensive and takes longer to compute, as well as being more complex in general [4]. Therefore, trajectory (classical) methods are preferred method of approach, and hence the importance in the understanding to its validity limits.

4.1.1 Limitations in classical Monte Carlo simulations in nanodosimetry

In this section I will examine the validity of trajectory methods. A key concept of quantum mechanics is the Heisenbeg’s uncertainty principle. This principle dictates that the product of
the de Broglie wavelength and uncertainty in the momentum of all particles should be equal to or less than a constant, $\hbar$, as shown in Eq. 4.4 [55]. Thus, it is expected that all particle simulators should satisfy Eq. 4.4 too. This forms the first of the two conditions needed.

$$\Delta x \times \Delta P_x \leq \hbar$$

(4.4)

The second condition which all simulations of particle transport must satisfy is related to $\Delta x$ and $\Delta p$ as shown in Eq. 4.5 and 4.6, where $s$ and $p$ represents the relevant length and moment in the simulation respectively. If any of the conditions outlined in Eq. 4.4-4.6 are violated for the simulated ionising particle then trajectory methods becomes inadequate to be used.

$$\Delta x \ll s$$

(4.5)

$$\Delta p \ll p$$

(4.6)

The de Broglie wavelength, $\lambda$, for particles of energies much less than $mc^2$, where $m$ and $c$ represents the mass of the particle and speed of light respectively (or also known as non-relativistic particles) is approximated as $\sqrt{1.50/E(eV)}$, it can be seen fairly easily that the de Broglie wavelength increases as the energy of the particle decreases. The de Broglie wavelength increases from 0.04 nm for a 1 KeV electron to 0.39 nm for an electron of energy 100 folds less at 10 eV. Ionisation path lengths for electrons of 100 eV in liquid water is approximately 1.5 nm, and for electrons of 1 keV energy the ionisation path length increases to approximately 3.8 nm. However, there is a slight dip in their linearity at around electron energy of 150 eV where the step length drops to 1.4 nm. The de Broglie wavelength becomes more comparable and even larger than the mean free length of liquid water the lower the energy the particle has. For instance, the ratio of the de Broglie wavelength over the mean free path $s$, $\lambda/s$, for a 100 eV
electron is \( \approx 24\% \) based from unit density mean cross section data for liquid water from work by Kawrakow [80], and similarly for water vapour unit density cross section data obtained from Nikjoo [114] the ratio is \( \approx 32\% \).

Let’s imagine a particle with energy \( E \) interacts with a medium by transferring energy of amount \( \Delta E \). During the interaction event, there is an uncertainty \( \Delta E \) associated with the energy transfer and an uncertainty \( \Delta x \) associated with the exact location of interaction. The relation between these two uncertainties are different for ultra-relativistic and non-relativistic particles. Here we will consider the general case where both cases will work.

Based from quantum uncertainty principle, Kaplan and Miterev [79] demonstrated that a particle of velocity \( v_x \) transferring energy \( \Delta E \) is related to its position uncertainty \( \Delta x \) by a constant. The exact formula is derived as follows:

Starting from relativistic mechanics the energy, \( E \), and momentum \( P_x \), of a particle with mass \( M_0 \) moving in direction \( x \) follow relations shown in Eq. 4.7 and 4.8.

\[
E^2 = c^2 (P_x^2 + M_0^2 c^2) \quad (4.7)
\]

\[
P_x^2 = \frac{E V_x}{c^2} \quad (4.8)
\]

Equation 4.7 can be rewritten as,

\[
E\Delta E = c^2 P_x \Delta P_x \quad (4.9)
\]

and upon rearranging, the above becomes:

\[
\Delta P_x = \left( \frac{E}{c^2 P_x} \right) \Delta E \quad (4.10)
\]

Substituting Eq. 4.10 into Eq. 4.4 and combining it with Eq. 4.8, we arrive at the final
expression for the spatial uncertainty, $\Delta x$, in the direction of motion for an interacting particle with velocity $v_x$ that is undergoing energy transfer of amount $\Delta E$.

$$\Delta x \geq \frac{\hbar v_x}{\Delta E} \quad (4.11)$$

The spatial uncertainty associated with an interaction site described in Eq. 4.11 can be applied for all velocity range, from relativistic to non-relativistic particles. As shown in Eq. 4.11, the uncertainty in location is inversely proportional to the energy lost. This means that the smaller the transfer of energy the greater the uncertainty in position. Whilst this may not cause a problem for high energy transfers such as in the ionisation of 1-s electrons where the position of the event can be relatively localised, it does become a problem when the energy transfer is too small. For most inelastic scattering processes the energy transferred are usually very small and thus the delocalisation in position can be significant. It is also the case for some of the most probable energy transfers for ionisation interactions, which is around $\Delta E = 15$ eV.

The boundary between the quantum and classical physics regime depends on the energy of the particle of interest. Typically the classical treatment of particles as point-like objects works fine in the 'high energy regime', while quantum mechanical treatment of particles are more applicable for the 'low energy' regime. The limit to the use of classical mechanics can be found through the following thought experiment:

Let $\Delta x$ and $\Delta p$ be the upper bound thresholds in uncertainties in position and momentum for the classical regime.

$$\Delta x = \epsilon s \quad (4.12)$$

$$\Delta p = \epsilon p \quad (4.13)$$

where $\epsilon$ represents a small and dimensionless quantity. Substituting the definitions for an upper bound on $\Delta x$ and $\Delta p$ in Eq. 4.12 and Eq. 4.13 into the Heisenberg’s uncertainty principle,
and approximating $E$ as $p^2/2m$, which is the non-relativistic approximation for kinetic energy. $m$ represents the particle’s mass. The combined equations consists of $\varepsilon_c$ - the critical value, the smallest possible ratio of uncertainties in position and momentum relative to appropriate length scale $s$, and the kinetic energy $E$ and some constants, as shown in Eq. 4.14. The classical treatments can be used within its validity only when the condition outlined in Eq. 4.5 as well as the requirements for $\Delta x$ and $\Delta p$ to be greater than $\varepsilon_c$ are all satisfied. These criteria are usually met by particles of interest during radiotherapy treatment planning as a typical energy cut-off is at $\approx 1$ keV and the relevant length scale $s \approx 1$ mm. This means that $\varepsilon_c$ is only just around $8 \times 10^{-5}$. However, these criteria may not be met for lower energy particle simulations such as nanodosimetric particle track simulations.

$$\varepsilon \geq \varepsilon_c = \sqrt{\frac{\hbar}{s\sqrt{2mE}}}$$  \hspace{2cm} (4.14)

The length scale of relevance in low energy - the range of concern for nanodosimetric simulations, is the average distance between interactions for a particle with kinetic energy $E$, known as the mean free path $s = 1/\Sigma(E)$. $\Sigma(E)$ is the average number of interactions per unit length, known as the cross section. Substituting the expression for $s$ into Eq. 4.14 we can arrive at an expression for the critical epsilon, $\varepsilon_c$, as shown in Eq. 4.15.

$$\varepsilon_c = \sqrt{\frac{\hbar\Sigma(E)}{\sqrt{2mE}}}$$  \hspace{2cm} (4.15)

As $\varepsilon_c$ is inversely proportional to the mass of the incident particle we see that the parameter $\varepsilon$ is small for heavy particles and big for light particles. This means that it is okay to use classical treatments of particles in simulations of heavy particles, however for small particles such as electrons and positrons $\varepsilon_c$ will be considerably larger which sets a lower energy threshold for classical treatments to be valid. Take an electron as an example, $\varepsilon_c$ is typically 0.01 for electron energy of 10 keV or higher, and in relation to the length scale $s$ this translates as 1% uncer-
tainty in both momentum and position. This means that the uncertainties satisfy Heisenberg’s principle and also $\epsilon_c$ is small enough to be neglected and satisfies the criteria for the application of classical methods. However, as the electron energy is reduced to 100 eV we see that $\epsilon_c$ increases from 5% at 1 keV to 17%-20% at 100 eV. This suggests that low energy electrons $\epsilon_c$ becomes large enough which means that the uncertainties $\Delta x, \Delta p$ do not satisfy the condition in Eq. 4.5. For most radiotherapy treatment planning the energy level of ionising particle beams is usually much greater than 1 keV and therefore there isn’t a need to consider the effect of uncertainties from quantum mechanics.

Interaction cross sections are different for different density and composition of particles in the interacting medium. The difference in interaction cross section thus changes the value of $\epsilon_c$. For incident electrons in air due to small interaction cross section $\epsilon_c$ is only less than 1%. This means that it is valid for applying classical simulations between electrons of ultra-low energies (less than a few eV) and air. Proportional counters, as will be discussed later, is one form of experimental nanodosimeter where the interaction chamber filled with tissue-equivalent gases and so simulations of proportional counters using classical treatments are valid. The interaction chambers for these proportional counters are typically on millimetre scales and by using appropriate density conversions it is thought that the measurements can be used to study track structures at micrometer or even nanometer scales in condensed matter [127, 85] However as $\epsilon_c$ for water and gaseous medium are so different, it is debatable that the measurements in gaseous environment can be well related to track structures at condensed nanometer levels. It should also be noted that the uncertainties in water cross section data are also quite large for electron energy of below 1 keV due to different model approximations [41]. Thus, parameter $\epsilon_c$ also has a large variance. Cross section data for electrons of energy 100 eV to 1 keV has uncertainties typically around 20% to 40% but may be larger, and for energy of 100 eV or below uncertainties are even greater. This sets a natural constraint to the accuracies.
of classical simulations that relies on cross section calculations [41, 114].

4.1.2 Circumstantial validity of the trajectory method

As we have seen in the previous section, trajectory simulations that relies on classical mechanics are not suitable for simulating low energy particles, such as electrons of energy below 1 keV. However, this is not the only view on the matter. Liljequist and Nikjoo have pointed out despite the fact that low energy electrons have no well-defined trajectory in condensed matter, there are, nevertheless, evidences to support the use of trajectory simulations [97]. They have called the new limit for the use of trajectory methods as circumstantial validity.

As mentioned earlier, the validity of classical methods for multiple elastic scattering of a particle is when the de Broglie wavelength of the traversing particle, \( \lambda \), is much less than the intermolecular spacing, \( d_{\text{nm}} \), of the medium, i.e. \( \lambda << d_{\text{nm}} \). The circumstantial validity requirement as proposed by Liljequist et al. relaxes the need for significant differences between the two values. Instead, the condition for validity of classical trajectory simulation simply replaces the significantly smaller-than sign to just a smaller-than sign, as shown in the inequality in Eq. 4.16 [94].

\[
\lambda < d_{\text{nm}} \tag{4.16}
\]

This relation is applicable to all particles given that the particles in the interaction medium (also known as scatters) are randomly located. The point scatterer model works very well for low energy neutrons under an absence of nuclear magnetic moment as the range of nuclear scattering potential is short. In liquid water, the elastic scattering of thermal neutrons of energy around 0.04 eV can be modelled using trajectory method as it satisfies the circumstantial validity relation in Eq. 4.16. Neutrons, on the other hand, with energies below 0.01 eV (also known as ultra-cold neutrons) should only be treated by complete quantum methods [16] be-
cause ultra-cold neutrons violates the circumstantial validity requirement in Eq. 4.16.

Liljequist et al. has shown two distinct evidences in support of their circumstantial validity condition, summarised as follows:

1. There has been good agreement between trajectory simulation and empirical data for secondary electron emission spectrum with solids [2]. Perhaps it is not completely a surprise seeing that model parameters in trajectory simulations are usually fitted from empirical results.

2. Good agreements with detailed trajectory simulations are also found with exact quantum calculations for plural and multiple electron elastic scattering events, where the de Broglie wavelength of incident electrons are comparable or smaller than the intermolecular spacing, \( d_{\text{nn}} \) [92, 93]. This means that good agreements are found for electrons of energy \( \approx 30 \) eV and even up to 15 eV. These simulations have also shown that the target medium can have the same values for density \( n \) and minimum inter-scatterer distance \( d_{\text{min}} \) as those of liquid water [93]. The interacting environment is assumed to be composed of randomly located point-scatterers, which occupies a large space with fixed boundaries. The particle source are generally either unidirectional [92], or from a spherically outward point-source in all directions [95].

Possible explanations for the observed agreements between simulated results using trajectory methods, empirical data and exact quantum calculations fulfilled by the circumstantial validity condition may be resulting from the averaging effect on coherent scatterings. As mentioned briefly in the second point above, scatterers are moving objects with random trajectory paths and no fixed position. The study on the effect of randomly moving scatterers in both classical and quantum simulating environments are interesting areas of research and are out of the scope of this work [77]. To my best knowledge, the boundary between quantum and
classical methods are not yet clearly agreed in the community, nonetheless, as seen by the two
points above the trajectory methods can still be circumstantially valid.

4.2 Important nanodosimetric quantities

As we have seen in the previous section, track structure simulations can be used to simulate
low energy particles provided that the circumstantial validity condition is met. In the following
sections I will work with trajectory simulations and examine how uncertainties in the interac-
tion location can affect important physical parameters in nanodosimetry.

All pathways can be categorised into two groups. The first group is called direct ionisation
and it accounts for about 35% of all the radiation-induced damages. The term direct damage
refers to the damage caused by the primary incident particle and not by any secondary or any
other particles that are produced as products from the primary particles’ interactions. Direct
damage depends mainly on the cellular chemistry in the target region [143], as well as on the
LET of the incident particle, though the latter is not seen to be significantly important [48].
During events involving direct energy depositions the targeted DNA molecule is likely to be
ionised or excited to produce lesions or breakage of chemical bonds [54]. The second group
is called “indirect damage” and it accounts for the remaining 65% of all damages. Secondary
species include free radicals and hydrated electrons. A major contributor towards DNA dam-
age is found to be OH• radicals, which are produced from primary interactions with liquid
water as we shall see soon [98]. Generally, radicals that are produced in close proximity with
DNA segments or are able to diffuse to biologically sensitive structures are particularly more
damaging.

As can be inferred from the name, indirect damages refer to interactions from interaction
products of the primary particle as it traverses along its path. As an example, an ionising
particle may ionise a water molecule and as a result produces a free electron and a H$_2$O$^+$ ion. The products, e$^-$ and H$_2$O$^+$ ion, will induce a chain of reactions with neighbouring molecules and continue to react until their energies drops below a certain threshold or are neutralised by other charged particles. The H$_2$O$^+$ ion is highly unstable and will inevitably break down into a H$^+$ ion and a OH$^*$ radical. The free electron is relatively less reactive to the ion and is likely to be captured by a H$^+$ ion or a water molecule to produce a H$^*$ radical or a H$_2$O$^-$ ion respectively. The H$_2$O$^-$ ion is also an unstable particle and will dissociate to a lower energy state: a H$^*$ radical and a OH$^-$ ion [3].

In terms of time scales for these interactions involving reactive species, about 10 ps after the initial primary particle’s interaction with water molecule the space surrounding the primary particle’s track will consist of H$^*$, OH$^*$ and free electrons $e_{aq}$ [99]. Other product ions such as H$^+$ and OH$^-$ ions are irrelevant towards radiobiological damages because they are found naturally in liquid water any ways at density of $\sim 1\mu$m$^{-3}$. They will be recombined to form neutralised aqueous water molecules should their concentration increase above water’s natural concentration. At up to a few nanoseconds after the initial event, if the free radicals have not reacted with anything else, they will randomly diffuse away from their initial site at a rate given by the concentration. They will continue to react with surrounding particles. If the target medium is biological tissue then possible biological entities which they will react with are proteins or DNA molecules, or even radical scavenger molecules that are found in cells to prevent biological damages. Typically, only those free radicals that manages to reach a DNA molecule without being neutralised by a radical scavenger are able to cause biological damages, and they accounts for only a small proportion out of the initial population.
4.2.1 Biological sensitive volumes

In computational nanodosimetry, a biologically-sensitive target volume in living cells is generally considered as a DNA segment of 10 base pairs. DNA, or Deoxyribonucleic acid in full, is the fundamental component of living cells that encode vital information for the survival of the cell. For ease of computation implementation, the dimensions of a segment of DNA strand of 10 base pairs is equivalent to a cylinder of 2.3 nm in diameter and 3.4 nm in height, and is filled and surrounded with liquid water. These dimensions correspond specifically to one convolution of B-DNA, which is one of three conformations of the repeating nucleotide chains in DNA that is found in living cells. It is common practice within the nanodosimetry community to use nanometric cylindrical volume that is equivalent to 10 base-pairs of the B-DNA as target volumes [110, 57, 88].

For an incident electron beam of 100 eV interacting with liquid water in a cylinder of equivalent dimension to ten base-pairs of DNA strands, possible interactions within the simulated volumes are elastic, excitation, ionisation, attachment and vibrational excitation. In fig. 4.1 we see that elastic interactions are the dominating interaction and is followed by vibrational excitation and ionisation interactions. Excitation and electron attachments are at the other end of the spectrum for being the least likely interactions in the target volume.

4.2.2 Categorisation of radiation-induced damages

Generally, the severity of damages caused by ionising particles on living cells in determined by the size and the density of the clustered ionisation event sites. For target dimension equivalent to DNA molecules of 10 base-pairs, there are three major classifications of damages induced by ionising particles:

Figure 4.1: For an electron beam of monochromatic energy of 100 eV in a cuboid world volume of liquid water of sides of 1 µm, this graph shows the number of events (y-axis) for all electron processes (x-axis), where the processes are integer values from 11 to 15. The corresponding processes are: 11 = electron elastic, 12 = electron excitation, 13 = electron ionisation, 14 = electron attachment, 15 = electron vibrational excitation.
Figure 4.2: Spatial locations of elastic interactions initiated by a beam of 100 eV electron. Spatial units are in nanometre.

Figure 4.3: Spatial locations of ionisation interactions initiated by a beam of 100 eV electron with liquid water.
Figure 4.4: Spatial locations of electron attachment interactions initiated by a beam of 100 eV electron with liquid water. Attachment events are represented as teal coloured dots. It is visible on a coloured electronic copy of this thesis if it difficult to see on the print.

Figure 4.5: Spatial locations of vibrational excitation interactions initiated by a beam of 100 eV electron with liquid water.

3. Base damage (BD).

The simplest form of DNA damage involves a single energy deposition event and is known as a Single-strand break, as shown in the first type classification listed above. The damage sites for these type of simple damages are usually on a backbone molecule of a DNA. They are one of the least effective cellular damage as they can be easily repaired by the cell’s repairing mechanisms. The other type of damage that is easily repaired by the cell’s repair mechanism is damage to base molecules of the DNA structure. Double-strand breaks refer to two single strand breaks that occur on different strands within a target volume equivalent of a DNA strand of 10 base pairs. There can be other scenarios of multiple damaged sites and depending on where the sites on relative to the DNA strand they can be: complex single-strand breaks, simple double-strand breaks, or complex double strand breaks [14], in the order of increasing complexity of damage. The more complex a damage is the more likely it is to induce carcinogenesis or cell death to the target cell.

4.2.3 Nanodosimetric parameters

An important parameter to characterise the biological effectiveness of ionising particle is the ionisation cluster size, $v$. This quantity is particularly relevant in nanodosimetry as it is defined as the number of ionisation events from interactions of a single incident particle within a biologically-equivalent and sensitive volume [56]. As discussed earlier, the common targets used for biological sensitive volumes are defined as a cylinder of diameter 2.3 nm and height of 3.4 nm. We shall adopt this definition of target volume in our works in the following section for consistency with the literature.

Ionisation cluster size is a stochastic quantity in nature. This means that for an incident
Figure 4.6: Classifications on DNA damage from ionising lesions

ionising particle of fixed energy and type, there isn’t a fixed corresponding ionisation cluster size. The non-determined nature is best described in a distribution function, $P(v|Q)$, where $Q$ describes the type and energy level of the incident ionising particle. We follow the convention notation in probability and so $P(v|T, Q)$ denotes the probability of having a cluster size $v$ given that the incident particle of energy $T$ has defined properties such as charge, mass and velocity. The ionisation cluster size distribution and its related parameters are shown to correlate well with biological experiments in terms of biological effectiveness [50, 51, 58, 105].

The mean number of ionisation events within a defined target volume is then a summation of all its possible cluster sizes from $v$ equals to 0 to infinity times a weighing factor that is by the probability $P(v|T, Q)$, as shown in Eq. 4.17.

$$M_1(T, G) = \sum_{v=0}^{\infty} vP(v|T, G)$$  \hspace{1em} (4.17)

The statistical variance on the probability distribution, $P(v|Q)$, is denoted as $V(T, G)$ and is
defined by Fano as the quotient of the second order of mean cluster size, $M_2$, with the mean cluster size, as shown in Eq. 4.18. $M_2$ is defined.

\[ V_{rel}(T; G) = \frac{M_2(T, G) - M_1^2(T, G)}{M_1(T, G)} \]  

where $M_2$ is defined as follows:

\[ M_2(T, G) = \sum_{v=0}^{\infty} v^2 P(v|T, G) \]  

The final important quantity in nanodosimetry that I will introduce is used to describe the complexity of the clustered ionising interactions and is defined as the sum of probabilities for having two or more ionisation events in the target, as shown in Eq. 4.20, $F_2(T)$. This addresses the more severe lesion scenarios as double-strand breaks require at least two or more ionisation events. It is shown that cumulative frequency, $F_2(T)$, is correlated with formation of double strand DNA breaks [60, 125].

\[ F_2(T) = \sum_{v=2}^{\infty} P(v|T; G) \]  

4.3 Effects of spatial delocalisations to ionisation cluster-size distribution

In previous sections I have described how trajectory simulations can still be used to simulate low energy ionising particle interactions with matter. In this section, I shall investigate how spatial uncertainties associated with low energy deposition events will affect one of the most important nanodosimetric parameter - ionisation cluster size distribution.

In my work, I have used a trajectory simulation package called GEANT4 (GEometry ANd Tracking)-DNA, release version 10.0.0, to investigate ionisation cluster size formations [89].
GEANT4-DNA utilises Monte Carlo computation methods and is described in more detail in section 4.3.1. Simulation procedures such as tracking and scoring are described in the following sub-section.

4.3.1 GEANT4 - a Monte Carlo method

A good overview on the early development on track trajectory simulations can be found on a paper written by Nikjoo et al. [115]. There are many track structure codes for particle interactions within the community, but they all produce slightly different results. The differences between them are due to the difference in their underlying cross section data and the implemented theoretical models [107]. At higher energy levels, the differences between different track structure codes can be neglected. However, at nanodosimetric (low) energy range their discrepancies becomes more significant. This is due to a lack of consensus in the cross section data in the low energy range. Typically, the cross section for low-energy electrons in water is obtained from water vapour experiments by applying appropriate density re-scaling. The differences between cross section data can be traced back to the different ways in which they are obtained, which include discrepancies in experimental cross-section results and differences in assumptions made in the conversion methods [137].

GEANT4-DNA is a simulation package for particle track structure using Monte Carlo methods from an ongoing research collaboration - GEANT4. It is originally initiated in the early 1990s by researchers at CERN and KEK as extension and improvement from GEANT3 and various other existing Monte Carlo codes [1]. GEANT4 collaboration was formed in 1999 and the initial aim of the working group was to create simulations of particle detectors used by complex nuclear physics experiments at NASA, typically for the simulation of high energetic particles. Thus, the original GEANT4 application specialised in high energy particle interactions. However, in more recent years due to the emerging need for particle simulators in the low energy...
range from the nanodosimetry community, GEANT4 is extended to include low energy physics lists. The new application is called GEANT4-DNA. Despite the inclusion of ‘DNA’ in its name, the default environment is only tailored for low energy particles interactions in liquid water environment, though there are other packages that includes DNA particles as the interacting medium. Due to the lack of model parameters for all low energy particles, GEANT4-DNA 10.0.0 only supports a limited number of interacting particles. Available interacting particles are electron, proton, carbon ion, oxygen ion, nitrogen ion, iron ion and hydrogen nucleus.

The are many physics models that one can choose to implement within a GEANT4 application. Different physics models differ in computation speed and different levels of physical approximations. For the low energy electron ionisation cluster distribution experiments outlined in this section, the physics models used are only associated with sub- 1 keV electrons [71, 47]. For electron elastic scattering interactions, screened Rutherford model is implemented with a low energy limit of 8.23 eV. Emfietzoglou model with low energy limit of 8.23 eV is used for electron excitation, and Born model is implemented for electron ionisation interactions with low energy limit of 12.61 eV. The high energy limit for all of these electron models are higher than 1 keV and thus covers the energy range of interest. GEANT4 can be considered as a semi-classical particle simulator. Despite the fact that all particles within the application are treated as point-like particles with definite energy-momentum and space-time position, the cross-section data and the computed final states of the particles mostly takes quantum effects into account. Errors in cross section data depends on the physical process. As summarised by Bernal et. al., in the low energy regime electron inelastic, electron inelastic excitation, electron excitation and electron ionisation cross section data in liquid water all shows error rate as high as 40 % in the 10 eV to 100 eV energy range [10]. For the purpose of this simulation experiment, these uncertainties are considered as systematic errors and does not affect the direct comparison between two applications written with the same intrinsic errors.
In terms of programming architecture, it adopts object-oriented concept and is fully written in C++ programming language. The implementation of object-oriented language was a novelty at the time as it was not of common practice as previous codes were usually not transparent and was difficult and complex to read and to re-develop. This allowed end-users to build effectively on top of the existing framework by importing relevant libraries and classes. An experienced user could even modify the physics processes used in models. The physics lists contained in every new releases of GEANT4 are the most up-to-date and complete repository for a wide energy range of particle-interaction physics. The GEANT4-DNA working group regularly updates new releases and is still active at present. Similarly, GEANT4 collaboration is also continuously expanding and most of their works are well-documented in the literature.

GEANT4 collaboration has also made the very important decision in making the codes publicly available for everyone. With the versatility and modularity that object-oriented programming practice brought, and together with the open-source nature of the project it was easier for everyone to contribute, modify and improve the codes. Further details on GEANT4-DNA applications can be found in the numerous publications by members of the GEANT4-DNA working group [71, 72, 24, 25, 142].

GEANT4 simulation package utilises Monte Carlo methods, which is a numerical method that is based on statistical approach to find solutions to differential equations [103]. It makes use of repeated sampling of pseudo-random number in an attempt to determine the behaviour of a probabilistic event. Differential equations for the transport of charged particle is usually very complex such that for most cases it becomes infeasible to solve when using traditional analytic methods. Numerical method can be used to solve complex differential problems and, in particular, Monte Carlo method is good for solving particle transport problems due to the stochastic nature of the transport problem.

In a GEANT4 application, basic aspect of the simulation environment such as detector
geometry, detector materials, particle generation, particle interaction, particle tracking and event storing and visualisation, are defined in a list of methods. However, out of the many instantiation classes there are three compulsory classes that an end-user must instantiate in order to build the application. These three mandatory classes are: G4UserDetectorConstruction, G4VUserPrimaryGeneratorAction and G4VUserPhysics.

Class G4UserDetectorConstruction is for the definition of system geometry. This includes all the components of the detector as well as its surrounding medium. Class G4VUserPrimaryGeneratorAction is for the initialisation of particle source. Class G4VUserPhysics is also very important in initiating the application as, by default, GEANT4 does not transport any particle without the user pre-registering what physics process(es) are required.

End-users can further define their applications by using another five non-mandatory user action classes. They are the intermediate scripts that links the user-defined settings to the main GEANT4 program. The following subsections encapsulates all the three compulsory and five optional base classes. A diagram of the hierarchy of the whole classes architecture is shown in Fig. 4.7.

Basic concepts in a GEANT4 application are summarised as below:

**Primary particle generator**

Primary particle generation in GEANT4 is instantiated by the mandatory user action class G4VUserPrimaryGeneratorAction. This basically defines the source of the incident particles. This class requires the definition of two instances, G4PrimaryVertex and G4PrimaryParticle. The former states the particles starting point in space and time. The latter states the primary particle’s type, initial momentum and other characteristics.
Particle Gun

The original method for generating particles in GEANT4, G4ParticleGun, is designed to simulate a beam of particles. The particle gun allows customisation of particle type, energy, polarisation, charge and number of particles shot per event. It is possible to affect the particle gun differently with random (or pre-defined) characteristics dependent on event number and other customisations.

General Particle Source

The G4GeneralParticleSource is a more advanced implementation of the particle generator [27]. It is included in the standard GEANT4 toolkit provides built-in support for spectral, spatial and angular distribution of generated primary particles. The General Particle Source is easily controlled within existing applications using a built-in command tree in the user interface.

Description of Physics

Class G4VUserPhysicsList is for the definition of all particles and their associated physics processes. Each processes can take user defined cut-off energy level, below which the particle is dropped off from further tracking. Common practice is to activate GEANT4-DNA related models or standard models from GEANT4 depending on the interaction region in relation to the sensitive detector region. The switching between implementation of different physics models is facilitated by class G4Region. The implemented physics lists are the same for every distinct G4region. In order to minimise the boundary effect between different regions, it is better to extend the more detailed physics list to slightly beyond the sensitive detector region.
Figure 4.7: Figure shows the top level diagram of GEANT4 simulation toolkit as adapted from Agostinelli et al. [1]. Open circles represent the end with higher level of dependencies in the adjoint nodes, where the lines represent the a relationship.
Geometry

Class G4VUserDetectorConstruction is for the general definition of user-defined geometry associated with detector and its surroundings. Useful attributes in this class are materials and geometric locations of each component detector sensitivity and visualisation components. There are two ways to define a volume object in GEANT4: Constructive Solid Geometry (CSG) method and Boundary Represented Solids (BREPs) method. The easier one to implement out of the two is CSG and is also usually faster to compute. However, BREPs allows the user to construct much more complex and non-conventional solids [5]. The GEANT4 package comes with a handful of pre-defined simple solid objects which becomes extremely useful when constructing the geometry using the CSG concept. The simplest building blocks are boxes, tubes, spheres, cones, wedges and toruses. More complex shapes such as trapezoids, paraboloids and tetrahedra, are also available and are documented on their official site.

Within a GEANT4 application it is also possible for the user to use different physics models within different parts of the simulation environment, which can be defined in the DetectorConstruction class. This option is typically used when stepping through a more accurate model becomes too computationally expensive and unnecessary for geometric regions that are of little importance, for instance regions outside of the sensitive detector region. It is generally of better coding practice to reduce the computational intensity at non-critical regions and it can also be achieved through dumping existing particles or generation of secondary particles of sufficiently low energies from the tracking list.

User-defined actions

There are a couple of useful optional classes for end-users to insert a pre-defined action, such as scoring and tracking of interested events.

- G4UserRunAction allows user to define user-specific actions at the instantiation and end
of each run.

- **G4UserEventAction** is similar to class **G4UserRunAction**, however instead of inserting user-defined actions at each run this class allows user to insert it at the beginning and end of each event. An event is defined as each interaction.

- **G4UserSteppingAction** is for defining what happens at every step.

- **G4UserStackingAction** controls the internal track stacks.

- **G4UserTrackingAction** allows user to insert custom-defined actions at the beginning and end of each track.

### 4.3.2 Simulation procedures

The target volume of interest in this work is chosen as a cylinder with diameter of 2.3 nm and height of 3.4 nm. This corresponds to a biological sensitive volume that is equivalent to a DNA segment of 10 base pairs. The target volume is located in the centre of a world volume, which is of much greater dimensions such that interactions outside of the world volume is negligible. Both the world volume and target volume are filled with liquid water as to mimic the cellular environment where the water content is the most abundant. Another reason for using liquid water is due to the limiting available physics lists for low energy electrons in GEANT4-DNA at the time of writing. Low energy electrons are projected from the centre of the target cylinder (at half-height point), and towards the positive direction on the axis of symmetry of the target.

Ionisation cluster-sizes are defined as the number of ionisation interactions produced by one primary particle as well as all its secondary ions within the target volume. In this work, ionisation cluster size distribution is collected firstly without the consideration of quantum effects from spatial delocalisations associated with interacting particles. This follows the strict definition of classical mechanics and interactions are tracked within well-defined target volume
boundaries. This is then compared with all the ionisation events when the spatial uncertainty associated with interacting particles are considered. This includes ionisations that even occur outside the defined target volume provided that their associated delocalised spatial range falls on or within the target region. As discussed earlier, the spatial delocalisation is calculated from Eq. 4.11 from the initial velocity and mass of the interacting particle. I will be referring to the ionisation distribution that permits counts of ionisation events given the delocalised spatial range of particle as 'HUP-cluster-size distributions' from now on.

### 4.3.3 Description of tracking and scoring

Within a GEANT4 application, typically an user can insert custom codes in three distinct instances to track custom quantities. The first instance is at the beginning and/or end of every ‘run’. This is defined as the start and end of a whole application, where within each run of the application there can be multiple instantiation of incident particles. Each run can be initiated from GUI or can be called and initiated by the application in the form of a text file. The second key phase within an application is the beginning and end of each incident primary particle, known as a ‘event’. Primary particles are queued in series in the simulation stack and thus they are simulated one after the other. The third place of possible intervention for tracking is at the individual interaction level, known as a ‘step’. A step records interactions for every primary particles as well as all their secondary products. In this report I have tracked all the particle interactions and appended their results at each run of the application to a ROOT format file. ROOT is a data acquisition and analytic application that is designed for handling large data sets efficiently. Each ROOT file for this application contains the following basic information on the interaction:

1. $x, y, z$ coordinates
2. Interaction process

3. Interacting particle type

4. Energy deposition

5. Uncertainty in velocity

6. Uncertainty in energy deposition

Beside the ROOT file, a further two more .txt file outputs are used to record the counts for the number of interactions that falls outside a given sensitive region, and detailed interaction information for all that lies within the uncertain region given by the Heisenberg’s uncertainty principle.

4.3.4 Results and discussion

On average, the spatial delocalisations for ionisation interactions from electrons of energy 100 eV is found as 0.2615 nm in the simulation, and for electron of energy 1 keV this becomes 0.726 nm. Whilst the average energy transfer associated with ionisation interactions at the two energy level increased from 12.55 eV to 13.73 eV for 100 eV and 1 keV electrons, the rate increase in the velocity of the particle between the two energy levels is relatively much greater. At 100 eV, the average velocity of the incident electron during ionisation interactions is 4.81 mm/ns, and this is found to be 14.33 mm/ns for 1 keV electron primary particles. Therefore, within the target volume of interest the spatial uncertainties is found to increase on average. Furthermore, as the electron’s mean free ionisation path lengths for energy of 100 eV and 1 keV are approximately 1.5 nm and 3.8 nm respectively, the simulated result confirms that the circumstantial validity criteria is satisfied.

The ionisation cluster-size distribution for electrons of energies between 100 eV and 1 keV
Figure 4.8: Comparison of ionisation cluster-size distributions between two positions of electron beam inside a DNA-equivalent volume: 1) at mid-height, 2) at one surface.

in liquid water within the target volume is shown in Fig. 4.9. The HUP-cluster-size distribution is also plotted on the same figure for comparison.

From the simulated results, it is found that for electron energy of 100 eV around 5% of all the ionisation events inside a HUP-allowed target volume are originated from the outside of a strict target volume. Out of the 5% of these events, a majority of them helped to increase the counted cluster-size by one, at 4.76%. Whilst the rest 0.2% and 0.01% of these events contributed towards count increase of two and three respectively. It is observed that the distribution profile for the conventional and HUP-allowed ionisation distributions are very similar, where the percentage difference between the two is below 5 % for cluster sizes below 4. There is an increasing error rate at larger cluster sizes and this is due to the amplification of small population errors as the sample size for large cluster sizes are orders of magnitude smaller in comparison to smaller cluster sizes.

From Fig. 4.9, the ionisation cluster-size distributions for electrons of initial energy 1 keV is found to be logarithmic and the difference in the comparison between the conventional and
HUP-allowed distributions are not very different. Since the ionisation mean free path is larger for 1 keV in comparison to 100 eV, it is expected that the incident electrons will transfer most of its energy out of the target volume.

The average cluster size for electrons of energy between 100 eV and 1 keV is shown in Fig. 4.10. Again, the distribution under conventional clear-cut boundaries is compared with the inclusion of HUP. It is observed that the percentage difference between the two distributions are typically below 3.5% and the shape of the conventional distribution is not affected by HUP. This is validated by the comparison of distribution of variances, where the discrepancy is fluctuating around 1.5%, as shown in Fig. 4.11. Similarly, $M_2$ is not seen to be perturbed by the inclusion of HUP, as seen in Fig. 4.12.

Good agreements are observed between the nanodosimetric distributions simulated under the conventional method and the literature [59, 88]. Slight difference are due to the use of different physics list.

### 4.4 Concluding remarks

In this chapter, I have demonstrated the problems associated with using trajectory particle simulations for low energy electrons and special care needs to be taken for these simulations. However, as demonstrated by the circumstantial validity limit I have simulated sub-1 keV electrons in liquid water using GEANT4-DNA, and have validated that ionisation interaction events for electron primary particles below 1 keV satisfies the circumstantial validity criteria. Important nanodosimetric parameters including ionisation cluster-size distribution and $M_2$ are not found to be perturbed with significance by spatial delocalisations due to relations in Heisenberg’s uncertainty principle.
Figure 4.9: For electrons of initial energies 100 eV and 1 keV, this figure shows their associated ionisation cluster-size probability distributions as well as their HUP-cluster-size distributions. Figure is reproduced from one of my papers [89].
Figure 4.10: This figure is the average ionisation cluster size formation for electrons of energy range 100 eV to 1 keV. Figure is reproduced from one of my papers [89].
Figure 4.11: Figure shows the relative variance for low energetic electrons of energy 100 eV to 1 keV. Figure is reproduced from one of my papers [89].
Figure 4.12: Figure shows the $F_2(T)$ distribution for electron energy of 100 eV to 1 keV. Figure is reproduced from one of my papers [89].
Chapter 5

Inductive Superconducting

Transition Edge Detector (ISTED)

5.1 Inductive Superconducting Transition Edge Detector

The device ISTED was first conceptualised at National Institute of Science and Standards (NIST, Boulder) [102], and further modified and developed at NPL. An ISTED device consists of three distinct parts:


3. A SQUID: flux to voltage transducer.

The key difference between ISTED and other cryogenic detector is that an ISTED uses a passive heat sensitive absorber, as opposed to the ones under electrical connections. The heat sensitive absorber consists of a thin, low thermal mass superconducting material. It is placed
in the centre of a SQUID loop, and the two are coupled together via magnetic flux, i.e. under a current biasing SQUID the $V_{\text{out}}$ responds to changes in the penetration depth of the superconducting absorber film. The use of the passive component eliminates unnecessary electrical connections and thus reduces the sources of noise due to Johnson noise or electrical impedance mismatching. In principle, an ISTED device can achieve a higher energy and a faster time resolution. With the performance of an ISTED only dependent on the thermal mass of the absorber, and its thermal coupling with the substrate the minimum detectable energy is calculated to be $10^{-25}$ J/Hz, at a response time of sub $\mu$s for a nano-sized ISTED device [65]. The optimum resolutions are achieved by reducing the geometrical dimensions of the device as the decrease in loop size reduces its loop inductance.

Transition temperature of the absorber film, $T_{c^\text{a}}$, is chosen to be lower than the $T_C$ of the SQUID, $T_{C}^S$. This is so that when the ISTED is operating at the film’s biasing temperature the SQUID can still be operated in its superconducting regime, where thermal fluctuations are reduced. The film’s transition temperature can be easily tuned by varying its thickness, whilst the $T_C$ of the SQUID can be controlled by the level of e-beam irradiations at its two junctions.
When an incident particle hits the absorber, assuming that all energy and momentum from the particle are thermalised into the absorber, the resulting temperature rise in the absorber causes a change in its penetration depth, $\lambda$, as seen schematically in Fig. 5.3. Consequently, the change in the inductive coupling with the absorber and SQUID can be measured as the voltage difference across the SQUID, as shown in Fig. 5.2. The change in SQUID’s read out voltage are the inter-dependences of the SQUID voltage with inductance of its loop, and the rate change of its loop inductance with respect to absorber’s penetration depth, and the relation between the absorber’s penetration depth with its temperature. These inter-dependences can be written mathematically in Eq. 5.1 as the first, second and third terms on the right hand side respectively.

$$\frac{dV}{dT} = \frac{dV}{dL} \frac{dL}{d\lambda} \frac{d\lambda}{dT}$$

(5.1)

Detailed theoretical elaborations on Eq. 5.1 can be found in works of Ling Hao and John Gallop [65, 64, 66].
Figure 5.3: The dependence of film thickness to the magnetic field penetration and current density characteristics.
5.1.1 A simple model of a ISTED

Unlike the design of a TES, ISTED’s thin-film absorber is not electrically connected. This means that it does not have a Joule heating component. The mechanism that restores its operating temperature relies on simply thermal link between itself and the cooling bath from the cryostat. The whole system consists of a series of thermal conductances $G$ and heat capacities $C$, as seen in Fig. 5.1.1. The cooling power to the film’s phonon system is provided by the cryostat and it is responsible for cooling the detector after the detector is raised out of equilibrium with a time constant $\tau$. Figure 5.1.1 is an simplified thermal model of the system where the dominant factor for determining the thermal dynamics of the system is considered. As seen in Fig. 5.1.1 it is sufficient to model the entire system in terms of one heat capacity $C$ and one thermal conductance $G$ between the electron system in the superconductive thin-film and phonon system of the substrate.

Mathematically, the rate of temperature change in the electronic system of a generic calorimeter at temperature $T$ with heat capacity $C$ can be expressed as a function of the sum of input and output power, with a constant of proportionality $C$, as shown in Eq. 5.2. In this notation, $P_{\text{cool}}$ and $P_{\text{incident}}$ are the powers associated with cooling from cryostat and heating from any incident particles or noise.

$$C \frac{dT}{dt} = -P_{\text{cool}} + P_{\text{incident}}$$  \hspace{1cm} (5.2)

The electron system of the film is coupled to the phonon system of the film’s lattice by a constant thermal conductance, $G$. The cooling power $P_{\text{cool}}$ can be further expressed in terms of the substrate temperature $T_{\text{sub}}$, which is maintained by the cryostat, as seen in Eq. 5.3. The variable exponent, $n$, generalises Eq. 5.3 to all systems. Ideally, $n$ can be measured and it is dependent on the system’s dominant thermal conductance.
Figure 5.4: Figure shows a complete thermal links in an ISTED.

Figure 5.5: Figure shows an simplified thermal model between different links in an ISTED.
\[ C \frac{dT}{dt} = -\kappa (T^n - T^n_{sub}) + P_{Joule} \] (5.3)

Under a steady-state temperature we see that the rate change in the system’s temperature should be zero, giving the condition \( T^n = T^n_{sub} \). Suppose the system is perturbed by an incoming particle, in which the temperature is raised by a small amount, \( T = T_0 + \Delta T \), where \( T_0 \) is the steady-state temperature and \( \Delta T \) is the small increase in temperature. Substitute the new expression of \( T \) with small perturbation \( \Delta T \) into Eq. 5.3, and expand to first order in \( \Delta T \) we get the following:

\[ C \frac{d\Delta T}{dt} = -\kappa (T^n - T^n_{sub}) - n\kappa T^{n-1} \Delta T \] (5.4)

As we have mentioned earlier, under equilibrium the temperature of the film \( T \) is the same as the temperature of the substrate \( T_{sub} \). This means that the first term in Eq. 5.4 can be dropped and the equation can be simplified and rearranged to Eq. 5.5.

\[ \frac{d\Delta T}{dt} = -\frac{1}{C} n\kappa T^{n-1} \Delta T \] (5.5)

This can be further simplified as all the terms apart from \( \frac{\Delta T}{C} \) is found to be thermal conductance, denoted by \( G \), as shown in Eq. 5.6.

\[ G = \frac{dP}{dT} = n\kappa T^{n-1} \] (5.6)

Finally substituting the expression for \( G \) into Eq. 5.5 we get:

\[ \frac{d\Delta T}{dt} = -\frac{G}{C} \Delta T \] (5.7)
Equation 5.7 is a simple first order equation and solutions are found easily through simple analytic as:

\[ \Delta T(t) = \Delta T_0 e^{-t/\tau} \] (5.8)

which is a simple exponential form with constant \( \tau \). This constant can be written in terms of heat capacity, \( C \), and thermal conductance \( G \), as shown below.

\[ \tau = \left( \frac{G}{C} \right)^{-1} \] (5.9)

\( \tau \) in the generalised solution shown in Eq. 5.8 is the intrinsic time constant for the relaxation time from perturbed temperature to equilibrium. It can be seen that the temperature decays exponentially in time.

As ISTED is intended to be used as a calorimeter, the temperature increase in the film is caused primarily by interactions with incoming particles. The final change in temperature in the film’s electron system actually depends on the efficiency of the energy transfer between the incident particle and the film’s electron system. Thus, if we were to include the efficiency of the energy transfer process and denote it by \( \varepsilon \) then the final representation of the solution becomes:

\[ \Delta T(t) = \frac{E_{\text{ion}} \varepsilon}{C} e^{-t/\tau} \] (5.10)

where \( E \) denotes the energy of the incident particle. The magnitude of the voltage readout of an ISTED is related to the energy of the incident ion, the heat capacity of the Carbon thin-film and its energy transfer efficiency to the electron system of the superconductive thin-film. As the cryostat is kept at a constant temperature the device will return to its original temperature with a characteristic time.
5.1.2 DC response and signal

Now, with the SQUID biased we can finalise our description of a particle absorption event. SQUID response gives us a direct window (with some gain factors) onto the inductance change in the absorber. Therefore, we see:

\[ \Delta V = \text{constant} \cdot \Delta T \]

\[ dV(t) = c(T) \cdot e^{-t/\tau} \]

(5.11)

For an energy absorption of 10 keV and assuming the total heat capacity of the absorber \( C_{\text{tot}} \) is 1 pJ/K, the temperature change in the system is around 1 mK, as \( \Delta T \approx \frac{E}{C_{\text{tot}}} \). Thus for the application of nanodosimeter, the energy resolution of the detector should ideally be in the range of 10s of eV, this requires the absorber to have a very low volume and small specific heat. It is also desirable to have a time decay constant \( \tau \) in the range of 10s of ns or smaller. As \( \tau \approx \frac{E}{C_{\text{tot}}} \), we can also calculate a rough thermal conductance between the absorber and the thermal bath.
Chapter 6

Optimising mutual inductance between SQUID and absorber using 3D-MLSI

As we have seen in chapter 3, a SQUID is an excellent transducer between magnetic flux and voltage, and I will demonstrate in this chapter that the geometric design of a SQUID plays an important role in optimisation of its sensitivity. The effect of varying its geometric design on its sensitivity can be found through series of experimental results, however, as this process is generally time consuming an easier way is to study the effect of geometry designs through computational simulations. In this chapter, I will use a simulation package called 3D-MLSI to investigate the optimum absorber size in relation to the SQUID loop which maximises the

\[1\]

The simulations and analysis presented in this chapter are solely my work. I have used a simulation package 3D-MLSI created by Dr. M. Khapaev.
SQUID’s flux-to-voltage output. Recall that the thin-film absorber is made from a superconducting thin-film and the inductive coupling between the absorber and SQUID is subject to changes in temperature. 3D-MLSI is developed by Dr. Mikhail Khapaev from Moscow State University [83] and it has built-in physics lists for thin-film superconducting materials.

6.1 3D-MLSI

The external magnetic field $B$ that is perpendicular to the plane of a SQUID can be calculated from the readout of the SQUID by considering energy conservation laws within the system [22, 67, 84, 82, 83]. The measured readout from a SQUID is the potential energy of the SQUID system and in order to convert this into a magnetic potential energy, one needs to start from an equation for the total energy of the superconducting thin film, which is shown in Eq. 6.1.

$$E = \frac{\mu_0}{2} \int V \cdot \lambda^2 \cdot J^2 dv + \frac{\mu_0}{8\pi} \int V \int V \frac{J(r) \cdot J(r')}{\|r - r'\|} drdr'$$  \hspace{1cm} (6.1)

The first term in Eq. 6.1 accounts for the magnetic contribution towards the total energy. This term is derived from a combination of London equations, Maxwells’ equations and the Biot-Savart law [22]. The second term accounts for the kinetic energy of all the non-dissipative Cooper pairs and quasi-particles in the superconductor, which is proportional to the square of the current density, $J$. London penetration depth for the superconductive material is denoted as $\lambda_L$. All integrals in Eq. 6.1 are set on geometric boundaries over the entire superconducting region.

From the equation for the total energy of the system, Eq. 6.1, 3D-MLSI converts it into an inductance matrix using relation shown in Eq. 6.2.

$$E = \frac{1}{2} L^T L$$  \hspace{1cm} (6.2)
Here, \( \mathbf{I} \) represents a vectorised entity of the circulating current, and \( \mathbf{L} \) denotes the inductance matrix. The inductance matrix is a matrix representation of all the mutual inductances between different components, where its row and column index denotes all the possible components. This means that the self-inductances of all objects in the system are found along its diagonal elements, and everywhere else denotes the mutual inductances between different components. Therefore, matrix \( \mathbf{L} \) is a symmetric matrix as all its off-diagonal elements are mutual inductances, and mutual inductances between object \( i \) and object \( j \) are the same because their indices are interchangeable, as shown in Eq. 6.3, where mutual component between component \( i \) and \( j \) are represented as \( M_{ij} \)

\[
M_{ij} = M_{ji} \tag{6.3}
\]

Equation 6.2 can be expanded to include all components within the system. Let the total number of components be \( N \) and the expansion of Eq. 6.2 then becomes the sum of contributions from every component, shown in Eq. 6.4. \( L_i \) and \( I_i \) in this equation denotes self-inductance and current term for component \( i \) respectively.

\[
E = \sum_{i=1}^{N} E_i = \sum_{i=1}^{N} \left[ \frac{1}{2} L_i I_i^2 + \frac{1}{2} (I_i M_{i1} I_1 + I_i M_{i2} I_2 + ... + I_i M_{iN} I_N) \right] \tag{6.4}
\]

There are a few ways to solve the current density term \( \mathbf{J} \) from Eq. 6.1. One approach is called variational technique and is based on minimisation of free energy [22, 67]. However, this method is computationally expensive as it takes a lot of computer memories and time [82]. A good alternative that reduces a lot of the computation load is by using finite-element modelling approach. Finite-element modelling approach enables computer to solve complex differential equations much more effectively. Due to its effectiveness at solving complex differential equations, finite-element modelling method is widely adopted for various other engineering problems, such as thermodynamics and stress analysis.
3D-MLSI is an example of a simulation package that specialises in solving the current density in thin-film superconductors by using the finite-element approach [84, 83].

Since 3D-MLSI is based on finite-element modelling, the simulation environment in 3D-MLSI consists of a finite-element mesh shaped as triangles and with sizes that can be adjusted by the user. Figure 6.1 shows an example of the mesh grids generated by 3DMLSI for a square and a square-shaped loop. The size of the mesh determines the spatial resolution of the calculations. When it is set too large, the calculation can be completed quickly but at a cost of the accuracy. However, when it is set too small the calculated becomes closer to the continuous case but it may become infeasible given certain available computation power and time. Trade-off between the mesh size and accuracy can be found fairly quickly with a few trials. The generation of mesh grids for all the conductors in a simulation forms the first step in a 3DMLSI computation, and it’s called the pre-processor step.

Current density, \( J \), is represented as a scalar potential function, which is subject to boundary conditions. This means that the continuous current function is transformed into a set of discrete values and is distributed at finite-sized discrete spatial points across the superconductor. At every spatial interval (mesh) the computation is initiated by a set of state parameters. The state parameters can be passed on from neighbouring spatial interval or are initialised with default values if it is the first spatial interval at time 0. After solutions are calculated from the given differential equations in the current mesh, the results are then passed onto its neighbours to initialise further computation in other spatial intervals. The process iterates until a final output condition is met. This computation step is the second step of a 3DMLSI simulation, and it’s called the numerical core.

The program is designed to be used on thin-film superconductors the currency density vector is pre-defined to have zero component in the z-direction. In practice, thin-film object are defined as an object with a z-component in its spatial geometry that is much smaller than its
Figure 6.1: Figure shows the triangular mesh grid generated by 3DMLSI for superconductive thin-film (inner square) and SQUID loop (outer square loop), shown as white lines.
x- and y- dimensions, and its thickness (z-component) of the object must also be in the same order of magnitude as its penetration depth. In the pre-processing step, the generated grid mesh is a 2D representation of the system of the thin-film conductors.

The third and final step to a 3DMLSI calculation involves an extraction of the inductance matrix from the calculated total energy of the system from the numerical core. We have seen the relationship between the total energy of the system and the inductance components in Eq. 6.4. The self-inductance of the system can be calculated as a special case of the energy equation in Eq. 6.4, where we can have all off-diagonal current components equal to zero by using appropriate boundary conditions. This means that the only non-zero current terms left are along the diagonal. Equation 6.5 shows the final relationship between self-inductance, energy of system and current component.

\[ L_i = \frac{2E}{I_i^2} \quad (6.5) \]

Similarly, mutual inductance can be obtained by setting the diagonal current components to zero, and the final equation is shown in Eq. 6.6.

\[ M_{ij} = \frac{2E - L_i I_j^2}{I_i I_j} \quad (6.6) \]

### 6.2 Simulation procedures

The simulated environment in 3D-MLSI consists of one square loop, one square thin-film and one circular coil. All the components are made from the superconducting material Niobium, as shown in Fig. 6.2. The inner square film is modelled as the absorber film and it is placed inside a square loop of 30 µm in outer length and width of 1 µm, and equal-distance from the inner edge of the square loop. Whilst the square loop represents the SQUID and is fixed in size.
the absorber film is of variable length. Josephson junctions are neglected in the simulations of SQUID as they are not the dominating factor in determining the mutual inductance between the SQUID and the absorber film. The outermost circular loop has a fixed inner diameter of 100 µm and loop width of 2µm. The purpose of the circular loop is to provide the system with an external magnetic field perpendicular to the SQUID plane.

When an ISTED is raised from its quiescent state by absorption of an incident particle, the rise in the absorber temperature results in a change in the inductance of the superconducting absorber film. Thus, I simulate the particle absorption event as a drop in the inductance in the absorber film. I then record the SQUID’s mutual inductance with the outer coil as we change the size of the absorber. With the outer length of the SQUID as 30 µm we repeat the simulations
with four different absorber lengths: 6 µm, 12 µm, 24 µm and 40 µm. For absorber length of 6 µm, 12 µm and 24 µm they are placed on the same plane as the SQUID as they are all small than the inner dimension of the SQUID loop and can all fit inside. On the other hand, the absorber with side 40 µm is larger than the inner SQUID loop and so it needs to be located at a different plane to the SQUID. In particular, the larger absorber film is raised on a plane that is 0.05 µm above the upper side of the SQUID film. In practice, the two layers can be electrically separated using an insulating layer, for instance, a thin deposition of polymethylmethacrylate (PMMA) spin-coating.

A 3D-MLSI program needs to be initiated with a .txt file and this consists of all the input commands to the program. In this .txt file the user should define all the necessary components in the simulated environment, using a 3D-MLSI specific language [81]. There are a few initialisation parameters for the program and they are geometric definition and location of all conductive components, global mesh size and London penetration depth $\lambda$ of all superconducting components. The SQUID and absorber components are both simulated with superconducting metal Niobium.

3D-MLSI allows user to define geometric properties associated with superconducting objects with simple pre-defined shapes such as lines, circles and arcs. Objects can also be placed at user defined positions using Euclidean geometry. Figure 6.3 shows a simple SQUID loop set-up in 3D-MLSI, along with an outer coil that completely encloses the SQUID loop for creating external magnetic fields perpendicular to the plane.

Niobium is characterised by London penetration depth $\lambda_d(T)$, which is a temperature-dependent parameter and for a given superconducting thin-film with given penetration depth at absolute zero temperature $\lambda_d(0)$ and transition temperature $T_c$, $\lambda_d(T)$ is found to follow Eq. 6.7. This means that for an operating temperature of roughly 7.5 K, London penetration depth for Niobium thin film is around 40 nm.
Figure 6.3: A schematic diagram of a SQUID loop generated by 3D-MLSI. The outer coil is used to generate external magnetic field.

\[
\lambda_L(T) = \lambda_L(0) \left( 1 - \left( \frac{T}{T_c} \right)^4 \right)^{-1/2} \tag{6.7}
\]

The first step in 3D-MLSI calculation involves decomposing the system components into appropriate triangular mesh grids, as shown in Fig. 6.4 and Fig. 6.5.

As mentioned earlier, the mesh grids define the boundaries for every individual finite elements within the calculation step. Figure 6.6, 6.7, 6.8 and 6.9 are examples of the final current flow through all the superconductors in the system. It can be seen that 3D-MLSI is able to generate current flow around the corners of the simulation components well.

### 6.3 Results and concluding remarks

Figure 6.10, 6.11, 6.12 and 6.13 shows the mutual inductance between SQUID and absorber film for varying absorber side of 6 μm, 12 μm, 24 μm and 40 μm respectively. It is found that for side
Figure 6.4: Generated mesh grids (shown as white lines) for system with absorber side of 6 µm.

Red lines represent geometric boundaries for each components.
Figure 6.5: Generated mesh grids (shown as white lines) for system with absorber side of 24 µm. Red lines represent geometric boundaries for each components.

Figure 6.6: Calculated current flow in the three conductor with absorber side of 6 µm.
Figure 6.7: Calculated current flow in the three conductor with absorber side of 12 µm.

Figure 6.8: Calculated current flow in the three conductor with absorber side of 24 µm.
Figure 6.9: Calculated current flow in the three conductor with absorber side of 40 μm. The absorber is shown to overlap the SQUID in this 2D bird-eye view of the system. However, the absorber is actually raised by 0.05 μm above the SQUID upper plane.
length of 6 \( \mu m \) and 12 \( \mu m \) the rate of change in mutual inductance with respect to absorber’s penetration depth is not significant enough establish a one-to-one relationship between the two variables. This means that for a change in temperature in the absorber the resulting change in the absorber’s penetration depth is not going to be detected by the SQUID due to the weak and indistinguishable mutual inductive coupling. This is perhaps not so surprising as the percentage of area coverage for these two absorber dimensions are only 4% and 17% with respect to the loop area. However, for absorber with side length of 24 \( \mu m \) and with area coverage of 68%, the rate change in mutual inductance varies is roughly quadratic. Similar quadratic characteristic is observed for absorber length of 40 \( \mu m \), moreover, the mutual inductance between the absorber and SQUID is shown to be stronger as compared with absorber with side of 24 \( \mu m \).

The quadratic behaviour between \( \lambda \) and \( M(SQUID, B) \) from Fig. 6.11 and 6.13 are due to the quadratic nature between current and mutual inductance, which can be seen from Eq. 6.6. As \( \lambda \) is proportional to the total amount of current flow in the SQUID it can be seen that increasing in \( \lambda \) should increases the mutual inductance quadratically.

In conclusion, the absorber film needs to cover at least roughly 68% of the SQUID loop area to produce detectable output in the SQUID and an absorber with side of 24 \( \mu m \) is large enough for sufficient inductive coupling with a SQUID loop of 30 \( \mu m \) in outer dimension.
Figure 6.10: For a square SQUID with side 6 µm, its mutual inductance is not observed to change significantly as the penetration depth of the absorber decreases from 0.04 µm to 0 µm.
Figure 6.11: At SQUID side of 12 µm, the mutual inductance is found to be more responsive to the change in absorber’s penetration depth as compared with the SQUID with side of 3 µm. However, the change in SQUID is still not significant enough.
Figure 6.12: There is a clear quadratic-like trend in SQUID’s response to changes in absorber’s penetration depth with side $24 \, \mu m$. This suggests that the bigger the absorber film the larger the output response from SQUID. However, as the simulated SQUID loop is only $30 \, \mu m$ across its inner loop side, any absorber film that’s larger than $24 \, \mu m$ will be challenging to fabricate using standard methods as the gap between the two components are approaching a few $\mu m$. 
Figure 6.13: By increasing the absorber side to 40 µm, the absorber no longer fits inside the SQUID loop and hence in the simulation it is suspended by 0.05 µm from the top of the SQUID plane.
Chapter 7

System Implementation and Device Fabrication

In this thesis, I have used three different methods of cooling systems for measurements of two different types of devices - nanoSQUIDs and microISTEDs. The cooling systems are chosen specifically to suit the need and purpose for each experiment. Generally, there is a trade-off between the mobility of the cooling system, available built-in external field strength and average time taken to load and un-load a device.

A 100 Litre liquid Helium dipping probe is used for an experiment to find the intrinsic noise level in the nanoSQUIDs (see later) because it is the designed housing cryostat for a SSA pre-amplifier. A smaller 20 L Helium cryostat with a 30 L liquid Nitrogen outer jacket is used for measurement on the ISTED, due to its small size for easy transportation between the cryogenic

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All the device outlined in this thesis are fabricated by Dr. David Cox. My sole contributions are in the experimentation and analysis of the RT characteristic of device E25 shown in this chapter. All other experimental works are achieved collaboratively.
lab and the $^{60}$Co irradiation room.

For the microISTED device I have measured its current-voltage (IV) characteristics near its $T_C$, resistance-temperature (RT) and its voltage response to $^{60}$Co irradiations. For the nanoSQUID device I have measured its IV, RT, voltage-flux ($V\phi$) and noise characteristics. Initial device characterisations (IVs and RTs) are carried out using pulse tube cooler as it is cyanogen-free and easier to assemble.

### 7.1 Cryostat Consideration

#### 7.1.1 Dipping probe

The liquid helium dipping probe used for the nanoSQUID measurements are fitted with two temperature stages, with one maintained at 4.2 K for SSA pre-amplifier operations and another at a user defined variable temperature.

#### 7.1.2 20 L liquid Helium dewar with liquid Nitrogen outer shell

The ISTED liquid helium cryostat system was already constructed by the group. Some key features includes the rf screened enclosure made of lead foil, lining the inside of a copper housing box for the device. The copper box is thermally connected with the base plate of the helium cryostat. The lead shielding is used to exclude electromagnetic waves using the Meissner effect from the superconducting lead walls. The device is assembled onto a sample stage (SQUID chip holder) which is fitted with a 50 $\Omega$ surface mounted resistor heater to provide local heating to the SQUID with stability of 1 mK in the range of 5 - 10 K; a 5-turn coil of diameter 2 mm, 2 mm above and 1 mm off central axis to the SQUID. Two diode based temperature sensors, Cernox, are thermally attached to the base plate and the sample stage. SQUID amplification is achieved with a room temperature dc amplifier with typical voltage sensitivity of 1 nV/Hz$^{1/2}$. 

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Figure 7.1: 100 L liquid Helium dipping probe with SSA probe inserted. Some readout devices used during intrinsic noise measurement of nanoSQUIDs are shown in the rack to the right.
Figure 7.2: The device window on the red dewar is facing the $^{60}$C irradiation source. The dewar is placed onto a work bench, which can be freely moved to adjust the irradiation position along the irradiation path.

Current is supplied from a Hewlett-Packard hp 3245A, whilst voltage readings are taken by digital multimeter at room temperature.

### 7.1.3 Pulse Tube Cooler

Pulse tube cooler extracts thermal energies from objects by coherent mechanical pulses of Helium gas. It is capable of reaching a, more than sufficient, base temperature of 2.8K. Given its low operational cost and ease of operation it has been chosen as the cooler for the initial device characterisations.

Since the cooler uses mechanical pulses, extra care should be taken to anchor down any loose or excess wires or other components firmly to the sample plate, as movable parts should be eliminated. This can be done using Kapton tapes. By taping down the wires one also improves the contacting surface area between the cooling stage and the sample stage, meaning that the temperature of the whole sample stage shows fast thermalisation with the cooling stage.
7.2 Experimental System Set-up

The experiments that require the use of liquid cryogens such as the ones involved with the dipping probe and the 20 L liquid Helium dewar should be conducted with extra care as liquid Nitrogen and liquid Helium have high latent heat of evaporation, and they pose many safety hazards. One potential risk in working with low temperature experiments is to avoid using materials that become brittle under low temperatures. Use equipments only made from materials that are known to work well under low temperatures such as stainless steel, especially for the transport siphon which makes direct contact with the cryogen.

Before cooling down the experimental dewar with liquid cryogens one must ensure that the dewar has absolutely no leakages and the dry compartment that holds the sample stage
Figure 7.4: The sample chamber is attached to the cooling plate inside the pulse tube cooler.

has been pumped to at least $1 \times 10^{-4}$ mbar or smaller. This step ensures that there is minimal condensation in the dry compartment during the cooling process.

The transfer of liquid Helium to a large dewar such as the 100 L experimental dewar for the SQUID’s noise experiment should always be conducted after a pre-cooling stage with liquid Nitrogen first. Liquid Nitrogen is at 77 K and this step helps to prevent the cryostat from having thermal shocks and uneven local cooling rates. Liquid Nitrogen supply at our lab comes are from storage vessels of 100 L or larger, and the dewar has self-pressurising mechanisms which helps with the transfer process. The liquid Nitrogen transfer can be done via a vacuum insulated flexible transfer tube and the liquid is pushed through the tube by inducing a pressure gradient from the Nitrogen dewar to the experimental dewar. One common way to increase the pressure inside the liquid Nitrogen dewar is by using the self-pressurising mechanism that is fitted onto the storage dewar. After the transfer of liquid Nitrogen one should monitor the
Figure 7.5: The inside of an aluminium and copper shielded chamber. The device chip is clamped down to the sample stage.
temperature of the sample stage and wait until the temperature stabilises at 77 K. This should take around an hour or more depending on the thermal mass of the dewar. If at this point the outside of the dewar feels very cold or that the pressure gauge reading of the sample compartment inside the dewar isn’t decreasing, it is a sign of vacuum leakage. It is advised to stop further cooling process and to investigate the source of the leakage. Furthermore, if the system doesn’t suffer from leakage problems then the liquid Nitrogen should be flushed out of the experimental dewar before the transfer of liquid Helium. In the case for the smaller 20 L dewar the liquid Nitrogen is filled into the dewar’s outer jacket and is to be remained there.

Liquid Helium transfer is slightly more technical than liquid Nitrogen transfer as it has an even lower latent heat of evaporation, and one typically adopts good transfer practices to reduce loss as it is more expensive. The transfer process works on the same principle as liquid Nitrogen transfer. However, this time the pressure is produced by pushing room temperature gas from a ‘bladder’ balloon attached to the storage dewar at one of its gas vent onto the surface of the liquid Helium. Due to the low latent heat of evaporation the induced perturbation is sufficient for the job. Note that the valve to the bladder should be switched off initially as the insertion of the transfer tube will create a lot of boil off of Helium gas and there’s a risk for the balloon to explode. It is a good idea to open the valve briefly at the beginning to allow some gas to be stored in the balloon, and then shut the valve until when one end of the transfer tube thermally stabilises in the cryogen. Helium gas has high enthalpy and it is a property that can one used to one’s advantage to reduce liquid Helium loss through the transfer process. This means that the cold gas inside the dewar is a good source of cooling. At the end of the liquid Helium storage dewar, the transfer tube can be lowered slowly to allow time for heat transfer from the tube to the cold gas. Whilst at the other end of the transfer tube, only insert this into the transfer tube entry point of the experimental dewar once you observe a jet of liquid Helium coming out at the end. Make sure to insert the tube to the bottom of the dewar as the
evaporated cold gas can still be utilised as a source of cooling power to the dewar. The rate of the transfer can be adjusted by controlling the pressure at the liquid Helium storage dewar. Furthermore, at the initial transfer stage the rate of transfer should be maintained at slow as this increases the contact time between the cold Helium gas and the dewar.

The pressure provided by squeezing the bladder balloon may be sufficient to initiate the transfer process, however, when the experimental dewar reaches 4.2 K and it starts to collect the cryogen increasing the transfer rate at this point will not only save time but also reduce the loss of liquid Helium through the transfer tube. Should more pressure is required then connecting the storage dewar to a pressurised Helium gas supply with a suitable pressure regulator is a good option.

In the rest of this section, I discuss the techniques used to reduce signal noise caused by thermal emf, impedance mismatching, RF (radio frequency) and other interferences. The sources of signal noise in any experiment can be categorised as either external or internal. External sources are from background fluctuations such as the magnetic fields or the pick-up of radiations from the atmospheres. On the other hand, internal noises arise from within the system setup, which is usually due to the electronics such as from the preamp or other electrical noisy apparatus. Thermal emf is significant for DC or low frequency AC measurements. It occurs as a result of the set-up of temperature gradients within the system, especially at the contacts between dissimilar metals. This type of noise is minimised by minimising the number of different materials from which connecting wires are made as well as minimising temperature differences between the inevitable material interconnections.

Impedance mismatching between the sample and the preamp can induce effective system noise. The reduction of these noises is achieved by an addition of a resistor in series with the preamp.

To reduce RF pick-up and other types of electromagnetic interferences, the system is en-
closed inside an electrostatic shielding from the metal cryostat. Similarly, co-axial cables are used for all wirings outside of the cryostat. Leads are passed through RC filters to eliminate RF interferences. Interference noises from the 50 Hz main power supply is reduced by reducing the ground-loop effect between different components. This means that a common ground is shared for all the co-axial earth and the cryostat. A common practice to reduce the inductance cross talk between wires in the system is to twist and bond the pairs to form a, commonly called, twisted pair.

7.3 Magnet design

NanoSQUID operations require large value of magnetic field bias relative to microSQUIDs. The voltage output of a SQUID can be optimised when it is biased with an offset flux as the change in flux periodicity during measurement can result in a large change of SQUID signal.

In this section I report the modelling on the specification of the solenoid required to produce a peak axial field of 2 T, for a 10 A current, given the constraints on the geometry of a hollow cylinder with inner radius $R_{\text{min}} = 55$ mm, outer radius $R_{\text{max}} = 75$ mm, and height $Z_{\text{max}} = 75$ mm. The proposed magnet will be conduction cooled using a pulse cooler in vacuum. Figure 7.6 illustrates the set-up of the coil.

The magnetic field distribution along the principal axis of the coil is calculated from Eq. 7.1, where $\mu_0$ is taken as the permeability of vacuum, $a$ is taken as the radius of the superconducting wire, $n = \frac{Z_{\text{max}}}{2a}$ represents the total number of turns in one layer of coil along the $z$ direction, $N = \frac{R_{\text{min}} - R_{\text{max}}}{2a}$ represents the total number of layers of coil.
Figure 7.6: a) The relation between the axial magnetic field strength and the z-position. To be within 90% of the maximum field strength, z is found to have a tolerance range of +/-18 mm.
b) An illustration of the multi-layered solenoid.

\[ B_z = \sum_{j=0}^{n} \sum_{i=0}^{N} \frac{\mu_0 I (R_{\text{min}} + 2ai)^2}{2 \left((R_{\text{min}} + 2ai)^2 + (H - z - 2ai)^2\right)^{3/2}} \]  \hspace{1cm} (7.1)  

\[ L = \sum_{j=0}^{n} 2\pi N (r + 2ai) \]  \hspace{1cm} (7.2)

Given the geometric constraints on the coils and the desired axial magnetic field required, we can see from Fig. 7.6 that the vertical displacement from the maximum field position is found to be +/- 18 mm, with a 90% tolerance. The total length of the superconducting wire required is calculated as 3.8 km (Eq. 7.2. However, the cost for such lengthy superconducting wire is quoted as 13 K from Oxford Instruments and it may be economically impractical to add this coil onto the existing measurement dewar. An alternative and feasible option to reach such high magnetic field is to use the Adiabatic Dilution Refrigerator (ADR). Although in view of the long term objective of the project, the ADR can only be used to characterise the nanoSQUIDs and is not suitably designed to be connected with the beam line. This is still a standing problem.
and will be investigated further in the next phase of the project.

7.4 Device fabrication

7.4.1 NanoSQUID junction fabrication

It is known that SQUIDs that are fabricated using conventional tri-layer techniques are quite susceptible to show hysteric effects in their current-voltage (I-V) characteristics, especially for SQUIDs with dimensions smaller than micrometers. In the context of current-voltage characteristic in SQUIDs, hysteresis refers to the non-reproducibility of the current-voltage relation within one complete sweep of current as it depends on the direction of current sweep. This means that the I-V relation measured with incremental increasing current is observed to be different from when the current is incrementally reversed. Therefore, hysteretic SQUIDs does not make an ideal sensor.

In order to prevent hysteresis in nanoSQUIDs, over the recent years between National Physical Laboratory (NPL) and University of Surrey we have developed a fabrication method to produce nanoSQUIDs with Dayem-bridge junctions [63]. From a fabrication perspective, micro-SQUIDs generally adopt a tri-layer layout, which is difficult to be implemented on nanoSQUIDs due to technical limitations. Fabricating Dayem-bridge type junctions are a more practical way of producing nano-sized tunnel junctions as they generally take less fabrication steps. Dayem-bridge junctions, like all tunnel junctions, are essentially weak links in the SQUID loop. Whilst in tri-layer tunnel junctions the weak links are in the form of the sandwiched material, in a Dayem-bridge junction it is simply a constriction in the superconducting loop. Dayem bridges can be fabricated using Focus Ion Beam (FIB) or Electron Beam Lithography (EBL) and are generally less than 100 nm in length.

The fabrication process for Dayem tunnel junctions for our devices involves a Focussed
Ion Beam (FIB) milling on a thin sputtered film Niobium (Nb). Niobium film is chosen as the superconducting material for the SQUID loop as it has a relatively high transition temperature ($T_c$) of 9 K when unpatterned, as well as for its small penetration depth of $\approx 40$ nm at $T = 0$ K. Unpatterned film thickness is usually around 100 to 150 nm and it is initially sputter deposited onto a Silicon substrate of around 5 $\mu$m wide using optical lithography. Gold bonding pads are also added around every device, which aids wire bonding during pre-measurement’s device assembly process.

The tunnel junctions are fabricated from a dual-beam FIB system in a two-stage process. Initially, a layer of amorphous Tungsten is deposited to cover the entire areas over the junctions. This layer of Tungsten act as masks for the subsequent milling process. The milling process then continuous with e-beam decomposition of Tungsten Hexacarbonyl ($W(CO)_6$) through an gas injection system (GIS) in the FIB system. The pre-milling masking process is better than direct milling method in two ways. Firstly, as the Nb thin film at the constricted tunnel section can be protected against Gallium ion implantation during the milling process. Secondly, the mask or sometimes called capping layer (as shown in Fig. 7.8) can be used as a stable shunt resistor that is conveniently in parallel with the microbridge. The shunt resistor provides the sufficient damping to the junctions in the normal state and thus preventing non-hysteretic current voltage characteristics in the SQUID. After depositing the Tungsten layer, the next step involves milling of the W-Nb layer to produce two nanobridges of dimensions 60-80 nm in both width and length using Gallium ion beams. As an end result, the transition temperatures $T_c$ of the newly milled nano-Dayem bridged junctions are reduced to approximately around 7.9 K.
Figure 7.7: SEM image of microlSTED. Courtesy of Dr David Cox

Figure 7.8: Illustrating the fabrication processes to create a nanobridge junction. Courtesy of Dr David Cox.
7.4.2 Thin-film absorber \( T_c \) tuning

The performance of an ISTED is optimised when it is most sensitive to changes in temperature. In order to be in the working range of the device, the superconductive thin-film absorber must have a transition temperature that is slightly lower than the \( T_c \) of the SQUID, typically the difference is around 1 K. This means that the absorber’s \( T_c \) is around 2 K below the \( T_c \) of pure Niobium film. Under this configuration, when the SQUID is operating at its optimum operating range the absorber is sitting just below its optimum working temperature. For the SQUID, its penetration depth \( \lambda \) is under the most sensitive region for a change in temperature. During an interaction event with an incoming particle, the thermal temperature of the absorber may be raised, which results in an increase in the SQUID’s penetration depth as well as changing the SQUID loop’s effective area.

In this section, I present two \( T_c \) tuning methods for the absorber film. The first method is through Gallium ion implantation by using focussed ion beam (FIB). In this method, a suitable density of Gallium ion exposure can be found by varying the energy and exposure time of Gallium ions to the Nb film until the processed film reaches a desirable \( T_c \). Depending on the energy level of Gallium ions used, this method is found to also change the thickness of Nb film as well as changing the film’s crystal lattice. The second method I have investigated is to tune \( T_c \) of Nb film directly by variations in its thickness during deposition processes. This method requires a final patterning step after the deposition process but may provide better control over \( T_c \).
7.4.3 Gallium ion dosing and SRIM modelling of film damage to irradiation

There are two possible interaction pathways between energetic ionising particles and a condensed material:

1. The energetic ion undergoes many inelastic collisions (such as ionisation) with the medium until its kinetic energy drop below a certain threshold and becomes relatively ‘inert’. These interactions cause damages to the internal crystal lattice structure of the condensed material and usually the level of damage is dependent on the depth from its surface.

2. If the incident ion has energy greater than the binding energy of surface atoms in the target medium then it is possible that the surface atoms will be released into the surrounding. This process in the semiconductor/material science community is called sputtering [10].

The sputtering process is generally facilitated by FIB with Gallium ion sources, and more recently Xe ions are more commonly used too, for it can be used to accelerate ions to sufficiently great energy.

In my studies, I adopt FIB in the tuning of $T_c$ on Nb thin films [13] using Gallium ion dosing techniques. Please note that all devices that are reported in this subsection are fabricated by Dr. David Cox and I was responsible for the measurement of films’ $T_c$.

The section of Nb film that is subject to ion beam exposure is roughly 3 $\mu$m in length along the full width of a straight track of 10 $\mu$m, as seen in Fig. 7.9. The range of ion doses are controlled using a 30 kV and 100 pA beam current in FIB. One unit (pass) of beam exposure is defined as a full sweep of a beam diameter of 23 nm through steps of 10 nm in both x and y direction (which accounts to 56.5% in spot overlap) and a beam dwell time of 1 $\mu$s at every step. On average 6.2 ions/nm$^2$ are delivered to the target area at every complete sweep of the
Figure 7.9: These images are scanning ion images of the Nb film at different stages of ion exposure to gallium ion beams, where the dashed yellow boxes represent the expore area. The beam is at 30 keV with current of 100 pA. It is observed that a total ion flux of $4.34 \times 10^9/\mu m^2$ is required to achieve total Nb film lift-off. Effects of ion channelling can also be seen. Left: Some microstructure is visible due to ion channelling in the grains of the film during the very early stages of the exposure. Middle: The film has lost half of its thickness but some unfavourably oriented grains have sputtered away completely while most still remain to form a continuous film. Right: Just before complete removal of the film some networks of the most favourably oriented grains still remain. Figure taken from one of my papers [90] (submitted and accepted).

beam. I will refer this one unit of ion beam exposure as one ‘pass’. The same pass are then repeated 25 times into a set, which equates to 155 ion/nm$^2$ per set. No more than 700 sets are performed on each device. Figure 7.9 shows a set of scanning ion microscopy images of the irradiated film with incremental exposures to Gallium ions. The Niobium films are measured to have total thickness of approximately 196 nm using FIB cross-sectioning. Simple lead and bonding pads are also made on the same chip as the Nb thin films using optical lithography techniques so that it is possible to conduct four-terminal $T_c$ measurements later.

Figure 7.10 shows the film’s RT response to some successively increasing dose of Ga irradiation. Further irradiation doses are carried out by Dr Sebastian Galer and an overall view of $T_c$ response to different Gallium ion exposure amounts are shown in Fig. 7.11. $T_c$ is observed to
Figure 7.10: R-T response General increase in the broadening of the transition in the dosed film is observed.

decrease from $\approx 8.2$ K to 0 K as the Gallium ion beam pass increases from 100 to 500.

There are many established ways to model ion implantation experiments in the semiconductor community. The levels of complexity in the existing simulations range from ion interactions at molecular level in specific orientations of single-crystal targets [148] to numerical modelling (such as ones that uses Monte Carlo method) in bulk material approximations, where ion doses are at the upper range that FIBs are able to handle ($\gg 19^2$ ions/$\mu$m$^2$). In the case of interest, as the number of interactions for the entire dosing event is large it is more appropriate for to apply ‘bulk’ material models as they are much quicker to run and do not need to track every individual interactions. SUSPRE is a popular simulation package for high energy ion beam
Figure 7.11: The transition temperature of the Nb film is observed to decrease polynomially with the increase of the number of Ga ion beam passes. The experiments are conducted on a batch of Nb film with the same thickness of $\approx 200$ nm. Figure taken from one of my papers [90] (submitted and accepted).
Figure 7.12: Calculated gallium implant profile for a 30 kV gallium ion dose of approximately 155 ions/nm² in niobium. Figure taken from one of my papers [90] (submitted and accepted).

Irradiation in condensed materials in the semiconductor research community, and is freely and publicly available. It is based from earlier theoretical works by Biersack [12] and is found to be in good agreement with another popular simulation package - SRIM. However, both SRIM and SUSPRE are approximations to the actual damage profiles as both neglects the filling of vacancies in the ion cascade. This forms a partial healing mechanism of crystal damage. At 30 keV and normal incidence, the expected sputter yield is found at 3.8 Nb atoms per Ga ion from SUSPRE calculations.

Figure 7.12 shows Gallium ion implant profile calculated from SUSPRE software after irradiation dose of 25 repeat passes. As can be seen on the prediction profile the peak concentration occurs at 12 nm from the sample surface with reaching Gallium ion implant concentration of roughly 15%. The Ga concentration becomes zero at a depth of 40 nm. In other words, the film is found to be fully amorphised after 100 repeat passes at depth 40 nm. The rate of decrease from the peak follows a Gaussian tail.

Experimentally, roughly 700 passes of Ga ions from FIB are needed to fully remove the
Nb film to expose the underlying Si substrate. This is equivalent to a milling rate of $\approx 0.28$ nm per pass of the beam assuming that the milling rate is kept as a constant. To put this into perspective, Niobium’s lattice parameter is 0.33 nm and so the milling rate per pass of the beam is almost at one atomic layer per pass. The actual sputter yield is 2.5 Nb atoms per Ga ion as Nb density is about 55 atoms/nm$^3$. SUSPRE prediction for removal rate is found to be 0.4 nm per pass given the calculated sputter yield and Ga exposure of 6.3 ion/nm$^2$. This means that the calculated rate is higher than the test result by about 40%.

One complication to the accuracy of SUSPRE model is that the target’s chemical composition is a dynamical variable, where the Ga concentration is expected to increase after every irradiation pass. The calculated sputter yield for Ga, Nb and the whole mixture from SUSPRE is shown in Fig. 7.13. It can be seen that the predicted sputter yield is consistently higher than the experimental findings of 2.5 atom/ion (on average) for all values of Nb/Ga content. The steady increase in sputter yield for Ga suggests, as shown in Fig. 7.13, suggests that the yield of Ga is expected to increase as the Ga concentration increases too during the milling process.

There significant discrepancy between the model and the experiment results may be due to the following two reasons. Firstly, the actual density of the Nb film may be lower than bulk Nb. If this hypothesis is correct then the stopping distance of the Ga ion will be longer than expected, meaning that some proportion of the irradiated Ga ion will penetrate deep enough into the target such that it can no longer cause sputtering. Secondly, the model may also have underestimated the importance of channelling effects that, similarly as the first reason, results in a proportion of the Ga ions to penetrate deeper (implantation) into the target material.

However, the film is unlikely to have a density that is of 65% of the bulk material as no abnormalities or porosity were observed in FIB cross-sectioning. This means that the first reason cannot be a plausible cause. Channelling effects, on the other hand, may be a more plausible explanation, as seen from the FIB images in Fig. 7.13. Channelling effects are generally de-
Figure 7.13: Calculated sputter yields from a 30 kV Ga ion for a mixture of niobium and gallium with increasing gallium content. Figure taken from one of my papers [90] (submitted and accepted).
pended on the angle of incidence from the irradiating beam. In a FIB system, this translates to the angle convergence of the beam. The beam is more diverged as the beam spot becomes larger at higher beam current. In our case, a beam current of 100 pA would produce a well-focused beam at the sample surface with little divergence. Therefore, the tunnelling effects are amplified if the orientation of the grains in the Nb film are favourably positioned for the incident beam. Thus, more Ga ions are transported deeper into the target material than prediction, which lowers Nb sputter yield. Therefore, the visible observation of the channelling effects in the FIB images supports the theory that more Ga ions are carried deeper into the material, and explaining why our measured sputter yield is considerably lower than the model.

Given the the average Nb removal rate discussed earlier, it can be seen that 200 passes of the dose would lift off a surface layer of 56 nm from the film. The irradiated film will still have a damaged layer of approximately 40 nm from calculations, which means 100 nm of the film should still be intact. From Fig. 7.10 we see the $T_c$ of the Nb film after 200 repeated irradiation passes decreases from above 8 K to below 8 K. As the film is passed by more doses of Ga irradiation we see the $T_c$ is suppressed accordingly. At around 350 repeat passes, the film should be around half of its original thickness with about 60 nm layer of still undamaged Nb. Beyond this dosage, the $T_c$ of the film is observed to drop more rapidly and eventually causing the film to exhibit no characteristics with 0 $T_c$. 


Chapter 8

Experimental results and nanobridge SQUID performance

1 In this chapter, measurements on the sensitivity-limit and basic characterisations of nanoSQUIDs that are fabricated using procedures outlined in chapter 7.4 are presented.

This chapter is organised into the following sections:

- SQUID current-voltage (IV) characteristics
- Background in noise - thermal noise in DC SQUID and flicker noise
- Experimental set-up
- SQUID noise measurement
- Optimise SQUID biasing points
- Experimental setup
- ISTED characterisations

1My contributions are in all of the experimental measurements and analysis.
8.1 SQUID IV characteristics

Current-voltage characteristics (I-V) for nanoSQUIDs have been measured at different temperatures. As example Fig. 8.1 shows an I-V curve at temperature 6.755 K. There are three distinct features to such IV curves: 1) the superconductive state is seen as a region of zero voltage before a certain threshold of current (critical current $I_c$), which indicates the DC Josephson effect, 2) the voltage jumps up like a delta function at the threshold current, which indicates AC Josephson effect, and finally 3) the I-V curve follows a linear relationship (e.g. the Ohms law) indicating that the superconductor makes the transition to normal state.

The IV characteristic of the nanoSQUID have been tested from operating temperature of 6.7505 K to 7.5 K as shown in Fig. 8.2. Note that the critical current of nanoSQUID increases with increasing bias temperature, and with decrease in temperature the slope of the AC effect part becomes steeper. The critical current as function of temperature has been plotted in Fig. 8.3. It can be seen that the optimum operating temperature for the SQUID can be as low as 6.7505 K, which makes it an excellent SQUID for the ISTED when coupled with an absorber film made using the $T_c$ tuning techniques described in section 7.4.2.

An I-V plot can be used to extract the optimum bias point to get steep V-$\Phi$ curve for NanoSQUID operation.

8.2 Background on experimental noise

The sensitivity-limit of any electronic devices are measures of their spectral power density. In this section, I will discuss the importance of spectral power density and why they are used in the noise analysis.

In an ideal scenario, assuming that we understand every underlying physics of an experiment noise signals can then be considered as any unanticipated signals. Noise signals can
Figure 8.1: IV characteristic of nanoSQUID at bias temperature of 6.755 K.

Figure 8.2: IV characteristics of nanoSQUID at temperature range from 6.7505 K to 7.5 K.
Figure 8.3: The change in critical current as a function of bias temperature for the nanoSQUID.
be generated either externally, independent of the experimental apparatus or internally, which arises from the materials that are used for the detector as well as any connections the detector is associated with. Mathematically, noise signals can be represented as a statistical fluctuation in time, which means that when a single measurement is repeated and measured to sufficient accuracy it is expected that the results for every individual experiments are slightly different. With sufficient repeats of an experiment, it can be observed that the readings are distributed with certain spread around its expected value, $y_{avg}(t)$. As denoted by the subscript ‘avg’ for average, $y_{avg}(t)$ can also be considered as the average or mean of the set of possible outputs in the measurement system in system output’s temporal domain, $y(t)$, and in Dirac’s braket notation it can be represented as $|y(t)\rangle$. All possible outputs are essentially a distribution with a mean and a variance, where variance is a measure of the spread of its mean, and is defined as the mean of least squared, as shown in Eq. 8.1 where $\sigma^2_y$ denotes variance. To put these concepts into perspective, the outputs from an ISTED measurement set-up are the SQUID’s DC voltage readings, and we should expect to see a certain spread in the readings even if the environment conditions are kept constant.

\[
\sigma^2_y = \int y^2(t) - 2y(t) dt
\]

\[
= |y(t)\rangle + |y^2(t)\rangle dt
\]

\[
= |(y(t) - y(t)^2)\rangle
\]

The term $|y(t)\rangle$ can be dropped from Eq. 8.1 as it can be offset to zero for simplicity, as shown in Eq. 8.2.

\[
\sigma^2_y = \int y^2(t) dt = |y(t)^2\rangle
\]

However, the equation for noise variance above is not enough to capture the whole complexity in noise. More concretely, the value of variance can be viewed as the ensemble average.
of outputs in an infinitely many and statistical identical systems, which neglects the significance of temporal relations between individual outputs. For instance, a perturbation in temperature in a material at time $t$ will have a consequence on its temperature at a subsequent time frame, $t + \Delta t$. This means that the temporal noise signal is actually an autocorrelation with itself, and in mathematical terms this means that the overall noise signal at time $t$ is a result of a convolution of its true input at $t$ with all noise signals in previous time frames, as shown as $R(t)$ in Eq. 8.3.

$$R(t) = \int y(t)y(t+t')dt = \langle y(t)y(t+t') \rangle$$ (8.3)

The autocorrelation function, $R(t)$, is still time dependent and so it is still a challenge to represent its underlying relations explicitly. The solution to the problem comes from Fourier transformation where the temporal series $R(t)$ is expressed in its frequency domain, $W(f)$. In the frequency domain, the noise signal is represented as a function of the frequency of occurrences for perturbation events, thus describing the correlation between these events. Fourier transformation from the temporal $R(t)$ to frequency domain $W(f)$ is done by an integration of the autocorrelation function with $e^{i\omega t}$ with respect to time, as seen in Eq. 8.4 where $\omega = 2\pi f$ is named as the angular frequency. $W(f)$ is also known as the spectral power density. This is an important term describing the noise characteristics of a system.

$$W(f) = FT[R(t)] = \int e^{i\omega t} R(t) dt$$ (8.4)

### 8.2.1 Thermal noise in DC SQUID

Thermal noise or Johnson noise is the dominant noise source to dc-SQUID with resistively shunted junctions. This type of noise is related to the operating temperature, SQUID’s inductance and capacitance in a relation shown in Eq. 8.5, where numerical constants $k_B$, $L$ and $C$ are
the Boltzmann’s constant, inductance and capacitance of the SQUID respectively. \( T \) denotes the operating temperature.

\[
S_B^{(1/2)} = (4k_BT\pi L/C)^{(1/2)} \tag{8.5}
\]

As the superconductor transits into its normal state when the biasing current exceeds critical current, \( I \gg I_0 \), the resistive shunt across the Josephson junctions will show an associated thermal noise current (also known as Nyquist noise) with a spectral density as shown in Eq. 8.6, where \( k_B \) denotes Boltzmann’s constant, and \( T \) and \( R \) have their usual meaning of temperature and resistance respectively.

\[
S_I(f) = \frac{4k_BT}{R} \tag{8.6}
\]

Nyquist noise can be observed by the following two features on an I-V characteristic of a superconducting device:

1. 'Noise rounding' - this is seen as a rounded I-V transition from superconductive to normal state. As a result of thermal noise at low voltage levels, the critical current can be observed to be lower than the actual value.

2. Current-induced voltage noise - this happens when the device transits into its normal state, i.e. when voltage is raised above zero, the noise is directly related to the current.

Noise rounding can be described rather nicely by the ‘washboard’ model which I have introduced previously in Chapter 3. In the case where \( I < I_c \) thermal noise, in effect, tilts the washboard potential, causing the total current to exceed the critical current. This extra tilt causes the particle to roll out of its current potential minimum to the next. For over-damped junctions the potential tilt produces a random time-series of voltage spikes. Thus, at currents
below the critical current the dc voltage becomes finite upon averaging in time. This is the origin to 'noise-rounding' and the reason why we will obtain a critical current at reduced value.

One way to categorise the significance of the thermal noise in the SQUID is to compare it with Josephson coupling energy. The noise is considered as a source of small fluctuations when both the ratio of thermal noise to Josephson coupling energy and the normalised inductance of SQUID are small, or in other words are much less than 1. The ratio of noise to Josephson coupling energy is known as the noise parameter. Similarly, the way to determine if the thermal noise source is significant or not is to see if the noise parameter is comparable to or greater than one.

Noise currents can be considered as a random distribution of Gaussian numbers [134]. The higher the noise parameter the more rounded and suppressed is the observed critical current, until the noise parameter becomes large enough (or if $I_C$ is small enough) that the thermal noise over-powers the Josephson coupling energy completely.

Therefore the required condition to achieve good SQUID performance is when Josephson coupling energy is much greater than thermal noise, as approximated in equation 8.7. The left hand of this equation is the Josephson coupling energy.

$$\frac{\Phi_0^2}{2L} >> 2\pi k_B T$$

(8.7)

The thermal or Nyquist noise that originates from the shunt resistors that we have discussed so far is typically a white noise. This means that it introduces a voltage noise evenly across all frequencies. The measurable voltage spectral density $S_V(f)$ can be expressed as flux noise spectral density by the conversion shown in equation 8.8.

$$S_\Phi(f) = \frac{S_V(f)}{V_\Phi^2}$$

(8.8)

Another useful parameter to quantify SQUID's noise spectrum is by writing it in terms of
its inductance. This is a good way to characterise and compare between different SQUIDs.

\[ \varepsilon(f) = \frac{S_\Phi(f)}{2L} \]  

(8.9)

### 8.2.2 Flicker (low frequency) noise

Low frequency noise, or also known as, Flicker noise or $1/f$ noise becomes a significant problems when the signal of observation occurs at frequency of 0.1 Hz or below. This usually have a greater impact to applications in specific areas such as biomagnetism and geophysics. As the name $1/f$ suggests the low frequency noise follows an inverse relationship to frequency. Thus, the noise level increases exponentially at lower frequencies.

### 8.3 SQUID noise measurement

As discussed in Chapter 3, a SQUID consists of two Josephson junctions connected in parallel in a loop of superconducting material. A SQUID is used as a very sensitive magnetometer typically of dimensions in the micron range. Best performance in SQUID is achieved using ultra-low transition temperature materials. However, temperatures below 4.2K (liquid Helium’s boiling point) requires expensive cooling refrigerators, and with the developments of dry cryogenic techniques that are relatively cheap and easy to use, materials such as Niobium with transition temperature above $T_c$ of liquid Helium is favourable.

Impedance matching between the amplifier and the device is an important practice for all detection and amplification applications. This requires both the impedances of the amplifier and the detection device to be within comparable range of each other. For the case of a nanoSQUID, it requires low impedance amplifier as itself is of low impedance and is one of the most sensitive detectors. This high specification on the amplifier rules out most common room
Figure 8.4: An example of the periodicity behaviour of V-Φ characteristic of a nanoSQUID where an external perpendicular magnetic field is supplied by a coil with current $I_{mag}$.

temperature amplifiers but low temperature devices such as SQUIDs are capable of operating at the white noise region when coupled inductively to the nanoSQUID. SQUIDs are ideal amplifiers for this purpose as its voltage readout can be tuned to vary linearly over a large range of applied field. The SQUID’s V-Φ characteristic can be used to determine the biasing current and usually this is found at a point just above its critical current at zero-field.

The gain in flux-to-voltage transformation for the SQUID amplifiers can be maximised if they are connected in series. This serialisation of SQUIDs also improves the performance of the amplifier by increasing its operation bandwidth as well as increasing its impedance, which makes it easier for electronic implementation purposes [75, 144]. These type of cryogenic amplifiers are known as SQUID Series Array (SSA).

The SQUID amplifier is typically operated under a flux-lock configuration as it is impossible to distinguish flux measurements greater than one flux quantum otherwise. As shown in Fig. 8.4, the screening current of a SQUID is periodic with increasing applied external magnetic field. The periodicity of this characteristic is one flux quantum. Thus, it becomes problematic if one needs to measure flux of more than one flux quantum. In the flux-lock configuration,
A Series-SQUID Array (SSA) consists of many SQUIDs connected in series and with appropriate scaling on feedback coils and more complicated electronics.

however, the SQUID is inductively coupled to a nearby coil where the coil is sent with the inverse signal from the SQUID. The magnetic fields generated by the coil keeps the SQUID under a constant flux, as illustrated in Fig 8.3. The voltage output from the SQUID at the constant flux state is a constant DC value, which is a benchmark for all other readings.

The SSA (cryogenic pre-amplifier) used in my experiments on nanoSQUIDs noise analysis are fabricated at Physikalisch-Technische Bundesanstalt (PTB), Germany, which required a constant operating temperature of 4.2K. The fixed operating temperature is achieved by having good thermal link between the SSA and the liquid Helium bath. The SSA signal is then fed into and processed by some Magnicon XXF FLL electronics before the final readout.

There is a heater attached to the sample stage of the nanoSQUID that allows the operating temperature of the nanoSQUID to vary from 6 to 8 K. The thermal links between the nanoSQUID stage and the liquid Helium bath is very weak and thus whilst the SSA is kept at
Figure 8.6: $V\Phi$ characteristic for 300nm loop nanoSQUID operated at bias temperature at 7.45K.

4.2 K the nanoSQUID can be maintained without perturbations at a desirable higher temperature. Furthermore, it is observed that the nanoSQUID stage can reach sub-mK stability.

8.4 Optimise SQUID biasing points

In order for SQUID to work at its optimum performance as a transducer of magnetic flux there are three parameters that needs to be fine-tuned: operating temperature, biasing current and biasing perpendicular magnetic field.

The flux-to-voltage transfer coefficient $V_{\Phi}$, and also written as $V_{\Phi}\delta\Phi$, can be a measure of the efficiency of the SQUID. The optimum performance of SQUID can be found at some given bias current and operating temperature that maximises the amplitude of the voltage modulation $V_{\Phi}$. The voltage change $\delta V = V_{\Phi}\delta\Phi$ in this regime is roughly linear. Typically, the external flux is found to approximate to $(2n+1)\Phi_0/4$, where $n$ denotes the set of all non-negative integers. At the optimum bias conditions it is possible for a nanoSQUID to detect
even a small flux change, e.g. when $\delta \Phi \ll \Phi_0$.

The maximum voltage change, $V_{\Phi,max}$, obtained from a flux variation of $\Delta \Phi$ up to $\Phi_0/2$ can be found using simple Ohm’s law considering that the resulting change in biasing current is $\Phi_0/L$. Total resistance of of the two parallel shunt resistors across SQUID junctions is $R/2$. As can be seen from equation 8.10, the maximum voltage drop is proportional to $R/L$.

$$\Delta V = R \cdot \Delta I = \frac{R}{2} \frac{\Phi_0}{L} V_{\Phi,max} \approx \frac{\Delta V}{\Delta \Phi} \approx \frac{R}{L}$$ (8.10)

Figure 8.7, 8.8, 8.9 and 8.4 shows examples of V-Φ characteristic of a nanoSQUID at biasing current of 0.1 mA to 1.3 mA (note that this is not the same device as the one presented in the noise analysis). V-Φ raw data are processed in Matlab; I have written a GUI (graphical user interface) application in Matlab to allow semi-automatic data processing, which helps to speed up the search and data acquisition process. The red lines represent tangents to the V-Φ curves and they are found as the steepest gradient of a parabolic fitting to the superconducting regions.
Figure 8.8: V-Φ characteristic at bias current of 0.15 mA.

Figure 8.9: V-Φ characteristic at bias current of 0.45 mA.
8.5 Experimental setup

Both the nanoSQUID stage and SSA are assembled onto an cryogenic insert which is made from a low thermal conductance and non-magnetic metal. Electrical connections to the nanoSQUIDs are made from superconducting wires in order to minimise Joules heating effects from conventional copper wires. An external magnetic field is applied normal to the plane of the nanoSQUID via an external solenoid.

The nanoSQUID is connected in series with a superconducting inductor and together they are in parallel with a resistor $R_B$ that is typically 0.1 $\Omega$, as shown in Fig. 8.10. When the bias current $I_B$ is less than the critical current of the SQUID $I_C$ all of $I_B$ goes through the SQUID’s branch completely. As the SSA is inductively coupled with the superconductive inductor the signal output $V_{out}$ from the SSA is the greatest in the case when $I_B < I_C$. However, as the bias current becomes greater than the SQUID’s critical current, i.e. $I_B > I_C$, the SQUID becomes resistive and causing some of $I_B$ to flow through the other branch with the resistor $R_B$. This then reduces the current through the superconductive coil and is detected as a drop in $V_{out}$ and the rate change of the voltage transfer function from the SSA.

$V_{out}$ from the SSA is measured as the power spectral density $S_v(f)$ of the nanoSQUID by a digital spectrum analyser. A final conversion from the power spectral density to the effective flux noise $S_{\phi}^{1/2}$ is done via Eq. 8.11, in which the two spectral densities are linked by the dc magnetic flux bias $V_\Phi$.

$$S_{\phi}^{1/2} = V_n \frac{\partial I_{SSA}}{\partial V_n} \frac{\partial \Phi}{\partial I_{SSA}}$$ (8.11)
Figure 8.10: Figure shows the schematic of the nanoSQUID and Series SQUID Array (SSA) circuit. The SSA is shown to pick up flux change from an inducer that is connected in series with the nanoSQUID. Voltage readout from the SSA is fed into a spectrum analyser for noise spectrum analysis. This figure is taken from one of my papers [119].
8.6 SQUID Noise analysis results and discussions

The current voltage characteristics for the nanoSQUIDs described in Chapter 7.4 follow closely the classic Resistively and Capacitively Shunted Junction Model (RCSJ). Figure 8.11(b) summarises the SQUID output voltage responses to variations in the biasing current and applied magnetic fields. The dimensionless critical current modulation parameter, \( \Delta I_C \), defined as the ratio of the maximum range in the critical currents over the maximum critical current (75 \( \mu A \)), \( \frac{\Delta I}{I_C_{\text{max}}} \), is greater than 0.78. The observed high value of the modulation depth suggests that the following two conditions are satisfied:

1. The first condition is that the McCumber shielding parameter, \( \beta_L = \frac{2I_C \Phi_0}{L} \), is less than 1.0. This implies that the upper bound on the SQUID loop inductance is 13 pH. This is in good agreement against the calculated loop inductance of 11.9 pH using 3D-MLSI software shown in Fig. 8.11 (a). It is interesting to note that the calculated geometric loop inductance accounts significantly for the total inductance. The good estimate from the geometric inductance for the SQUIDs Dayem junction suggests that the kinetic inductance contribution, which acts to reduce the SQUID response, is negligible under these conditions.

2. The second condition is that the critical current difference between the individual junctions, \( \Delta I = \frac{|I_{C1} - I_{C2}|}{I_{C1} + I_{C2}} \), must be less than 0.22. This implies that the two junctions are well matched, as a result from the precise and reproducible FIB milling of the junctions, and the good quality Nb thin film; i.e., uniformity, and in particular, an absence of grain boundaries within the junctions.

The intrinsic white noise associated with the Dayem bridge junction SQUIDs of dimension 65 nm x 65nm (Fig. 8.11(a)) has been measured as \( \approx 170 \, n\Phi_0 \) at 4.7 K (Fig. 8.12). This is an adequate level for the detection of a single electronic spin flip in a 1 Hz bandwidth. The
Figure 8.11: Figure (a) shows a single-electron microscopy (SEM) image of the nanoSQUID of loop diameter 200 nm and Dayem-type Josephson junctions of dimensions less than 60 nm. Figure (b) shows the screening current of the nanoSQUID as simulated from 3DMLSI. The self inductance of the device is calculated as 11.2 pH. Figure (c) shows the entire family of nanoSQUID's voltage response under a large range of bias current and applied flux. All figures are taken from one of my papers [119].
inductance of the SQUID loop has been calculated with software package 3D-MLSI to be 1.3 pH (Fig. 8.11). The energy sensitivity can be calculated as $4.6 \times 10^{-33} \text{ J/Hz}$ or around $45\bar{h}$, using equation 8.12, with substitution of $S = 3 \times 10^{-14} \Phi_0^2$ as the white noise level and the calculated inductance. The device is more than capable for our purposes as a nanocalorimeter, however, further developments to improve its energy sensitivity towards the standard zero point energy fluctuation for future purposes is required.

$$\varepsilon_n = \frac{S \Phi}{2L}$$  \hspace{1cm} (8.12)

Figure 8.13 is the result on the noise spectrum for a typical nanoSQUID that is shown in Fig. 8.11(a). The data presented here are the intrinsic noise of the nanoSQUID corrected for the noise in the preamp (SSA) during a zero current nanoSQUID bias. This means that the measured data are subtracted from the zero bias points following the conventional way. It is observed that the
noise difference between the biased and unbiased states becomes indistinguishable below a frequency about 100 Hz, thus it is not possible to perform the quadrature subtraction. For low frequency noise, where 1/f noise is dominant, the limitation on the readout by the SSA suggest that the Dayem junction nanoSQUIDs outperforms the trilayer junction based SSA. It may be because of an absence of two level noises in the nanobridge junctions.

Overall, the measured nanoSQUID exhibits a magnetic flux noise of $3 \times 10^{-14} \Phi_0^2$ in the white noise region, which is orders of magnitude more than capable of detecting energy depositions of sub-1 keV electrons. This suggests that the current design of SQUID satisfies the requirements as part of the calorimeter to the application of nanodosimetric measurements.

### 8.7 ISTED characterisations

Three identical ISTEDs are fabricated onto one single Silicon wafer. However in practice slight variations in the performances of each device are expected due to uncontrollable factors during the fabrication process; such as grain boundary defects in the wafer, or random ion implanta-
Figure 8.14: The RT characterisation on the three ISTEDs. Devices C30 and E20 are clearly more responsive to changes in temperature than device D25.

Prior to irradiating the ISTEDs with ion beams, examinations on the current-voltage characteristics (IVC) and the Resistance versus Temperature (RT) relations for the three devices are presented in this section. Moreover, the operation (biasing) conditions that optimise the device sensitivity are found and compared. The three devices are given the names D25, C30 and E20.

The RT characteristics of the three devices are collected under bias current of $5\mu\text{A}$, shown in Fig. 8.14. It is clearly seen that the SQUID transition temperatures for devices E20 and C30 are measured to be much higher than the one for device D25.

The IV characteristic for the three devices have been measured under zero field bias and are plotted in Fig. 8.15, 8.16 and 8.17 respectively. Device E20 in particular shows sharp junction transition at $I_C$ and a pair of well symmetric $I_C$s for both the positive and negative values. This suggests that the two SQUID junction $I_C$s are within close proximity and are both well resistively shunted.

The dependence of critical current with temperature of the devices are surveyed under zero field bias, shown in Fig. 8.18. The rate change of critical to temperature are plotted in Fig. 8.19.
Figure 8.15: The I-V characterisation for ISTED device D25 at temperature at 5.59K.
Figure 8.16: The I-V characterisation for ISTED device C30.
Figure 8.17: The I-V characterisation for ISTED device E20.
Figure 8.18: Critical current variations with temperature under zero field bias for three devices.

Equation of quadratically best fitted lines for device D25: \( I(T) = 6.8 \times 10^3 T^2 - 7.6 \times 10^4 T + 2.1 \times 10^5 \), E20: \( I(T) = 2.6 \times 10^5 T^2 - 4.56 \times 10^6 T + 2.0 \times 10^7 \), C30: \( I(T) = 3.7 \times 10^5 T^2 - 6.4 \times 10^6 T + 2.8 \times 10^7 \).

Figure 8.19: E20: \( \frac{dI}{dT} = 5.2 \times 10^5 T - 4.56 \times 10^6 \). C30: \( \frac{dI}{dT} = 7.4 \times 10^5 T - 6.48 \times 10^6 \).
Apart from SQUIDs characterisations the transition temperature of the absorber film has also been found, shown in Fig. 8.20. This film is measured on a separate track where it is deposited between Niobium electrodes. Using four point terminals, and fine tuning the bias current and the sampling rate of the measurements the absorber transition is observed, at 7.0 K. This verified that the transition temperature of the absorber is indeed just lower than the SQUIDs, which is at 8.55 K.

In conclusion, SQUID D25 is the least sensitive device out of the three. It can be seen from Fig. 8.14 that the critical current in SQUID E20 and C30 respond sharply to changes in temperature. It is calculated that when the devices are biased at the most sensitive temperature (when the slope of Ic vs T is the greatest), SQUID E20 can resolve a temperature step of 5 mK. This suggests that Ic measurements during irradiation can also be used to indicate thermal energies. In order to implement this the existing Labview programs has been modified to measure IV continuously in time, minimising the waiting time between each run.

Figure 8.20: RT of the absorber track.
8.8 ISTED under $^{60}$Co irradiation

ISTED E20 and C30 are tested under irradiation from a $^{60}$Co source. The $^{60}$Co gamma-ray facility used in this experiment is from a Theratron Radiotherapy irradiator maintained by the Radiation Dosimetry group at NPL. The $^{60}$Co source is rated at 300 TBq and the facility has a slidable work bench which can hold the experimental dewar at up to 6 m from the source.

The aim of this experiment is to test ISTED along with its cryogenic dewar and the measurement apparatus under a broad beam irradiation. One way to test ISTED with radiation is to align a particle beam with beam diameter that is equal to or smaller than the absorber film. However, given that the nano-beam facility at Surrey Ion Centre is still under progress and it isn’t clear that it can be completed before the end of my PhD program, the purpose of this experiment is to establish an alternative testing source for the ISTED. Broad beams will most certainly irradiate the whole area of the ISTED as well as its surrounding areas, however, with the correct particle energy and fluence rate the only part in the entire experimental system that is sensitive to the incident particles from the broad beam source should be the ISTED. The area covering the entire sample stage and the device can be considered to be well thermally coupled to a large heat sink.

For this experiment, the experimental dewar is placed 6 m from the Colbalt-60 source. Given that the activity of the source $S$ is 300 TBq and assuming that the irradiated particles are homogeneously emitted from the source in every direction, the fluence rate can be calculated as $\frac{S \cdot y}{4\pi r^2}$, where $r$ is the distance away from the source and $y$ represents the number of particles emitted for every disintegration of the radionuclide (yield). Colbalt-60 undergoes $\beta^-$ decay to produce a nuclear excited state of Nickel$^{\ast}$-60, an electron and an electron anti-neutrino. The electrons produced can have energies of 0.31 MeV with transition probability of 99.88% and 1.48 MeV with probability of 0.12%. The nuclear excited Nickel$^{\ast}$-60 from both pathways then
transits to the ground and stable state of Nickel-60. For the first pathway where the product electron has energy of 0.31 MeV, the transition from Nickel$^\ast$-60 to stable Nickel-60 produces two gamma rays of energy 1.1732 MeV and 1.3325 MeV. Whereas for the second pathway, the Nickel$^\ast$-60 transition produces a single gamma photon of 1.3325 MeV. Therefore, if we neglect the second pathway as it’s relatively too unprobable, the number of particles produced per disintegration of Cobalt-60 nucleus is three: one 0.31 MeV electron, one 1.1732 MeV photon and one 1.3325 MeV photon. This means that for our ISTED’s absorber of dimensions 800 nm by 800 nm, the fluence rate through the absorber is approximately $\frac{300 \times 10^{12} \times 3 \times 800^2 \times 10^{-18}}{4\pi \cdot 6^2} \approx 1 \text{s}^{-1}$.

A Labview program is specifically developed for fast I-V and R-T data acquisition required during the irradiation experiments. Input signals are collected via National Instrument 6281 which is a multichannel DAQ, capable of sampling rates of 18 bit at 500 KS/s.

RT characteristic of the device E20 are collected for pre- and post-$^{60}$Co irradiation, shown in Fig. 8.21. $T_c$ is observed to shift to a higher temperature after the irradiation, from 8.75 K to 8.77 K. The SQUID had been warmed to room temperature in between the pre- and post-irradiation measurements. Since the $^{60}$Co irradiated indiscriminately towards the device cryostat the change in SQUID’s $I_c$ suggests a permanent damage is developed in the SQUID junctions or superconducting materials.

The emitted particles (0.31 MeV electron, 1.17 MeV and 1.33e MeV gamma photons) from the $^{60}$Cobalt source can be detected by the sudden increase in temperature from a Cernox sensor. As the heater was set at a given temperature throughout the experiment it could be seen that the temperature is slowly reduced towards the set temperature from the equilibrated irradiation temperature. The voltage reading lagged the temperature reading and seemed to increase abruptly in quantised steps. As the radiation was turned off, sudden drop in temperature was observed, and followed by sudden drops in $V_{out}$.

The ‘quantised’ steps in voltage output during irradiation can also be seen in its I-V charac-
Figure 8.21: RT response from before and after $^{60}$Co irradiation. The device has been warmed up between the two measurements.

The characteristic prior to irradiation, shown in Fig. 8.22. The steps are likely to be caused by impurities or grain boundaries in the superconducting film in the SQUID loop, which is an artefact from device fabrication. Furthermore, it follows that during irradiation the SQUID is seen to detect a constant increase in temperature where the $\Delta T$ corresponds to a $\Delta V$ of 15 $\mu V$ at a constant 300 $\mu A$ bias current.

Figure 8.24 shows the SQUID and Cernox response to irradiation with more details. The SQUID lagged behind the temperature sensor by about 5 s and only responded in steps of 5$\mu V$. The Cernox temperature sensor is many orders of magnitude larger than the ISTED and therefore it is not surprising that it is more likely to respond to incoming particles. The SQUID is showing two distinct different rates of response. It responds to the incident particles with an initial fast response which is quickly followed by a slower one, which as discussed above may be from artefact from the SQUID loop due to impurities.

To achieve these SQUID responses it is found that the bias current needs to be carefully adjusted. The resolution of the voltage readout using Keithley 2635B sourcemeter is 100 nV.
Figure 8.22: IV characteristic of E20 before irradiation taken at bias temperature of 8.741 K. It can be seen that the SQUID in its normal state is showing a non-linear but fairly ‘stepped’ I-V relationship. This pattern is also seen during the irradiation experiments as shown in Fig. 8.23.
Figure 8.23: Voltage response from $^{60}$Co irradiation. The radiation source is directed towards the device with on and off intervals. Both the temperature response from the Cernox sensor and from SQUID is recorded.
Similar response of increased SQUID transition temperature after being irradiated is observed with device C30. In Fig. 8.27 we see that the $I_c$ of SQUID C30 has increases by 10 mK after irradiation, which is measured after a warming up to room temperature. However, during the irradiation process the $I_c$ is observed to decrease with increasing dosage. Different dosages from the same $^{60}$Co source is achieved with the addition of a wall of lead, which attenuates the source power by a factor of 10. Figure 8.28 shows the results of more repeats of RT at the two different dosages. Higher dosages, RT2,1 and RT4,1, shows a smaller drift than the lower dose (the rest of the data sets), which is counter-intuitive. However, since the higher dosed RTs are taken before the lower dosed ones we may expect damages in the SQUID to accumulates onto each new run. This is clearly observed from RT8,1 to RT16,1 as the transition temperature gradually decreases with consecutive runs. However, the device’s transition temperature is observed to shift down as opposed to up, and it is likely to be caused by a measurement artefact.

Figure 8.24: Voltage response from $^{60}$Co irradiation.

with response time of 1.2 ms.
Figure 8.25: Voltage response from $^{60}$Co irradiation.

from the self-regulating temperature sensor on the sample stage. As the RT measurements are taken with a small temporal step during constant irradiation, the sample stage heater would be adjusted to increase the cooling power in the latter instance. Thus, the measured temperature at the latter instance suffers from a systematic under-measurements on temperature.

It is also noted that the critical current of SQUID C30 after irradiation has shown an general increase with respect to temperature (Fig. 8.26). It is also found that the transition temperature of the device is suppressed during irradiation, as shown in Fig. 8.27 and 8.28 respectively. However, upon thermal warming the transition temperature is raised back to its value at pre-irradiation, which may be due to some thermal cycling mechanisms.

Overall, these sets of irradiation experiment with Colbalt-60 source are useful reference for future irradiation experiments:

- It is shown that the current device fabrication techniques are good enough to produce ISTEDs that are unperturbed by prolonged irradiation from a Cobalt-60 source. This is
an important finding as it means that the device can be used repeatedly without degra-
dation.

- Following from the point above, the entire measurement system is also robust enough
  against Cobalt-60 irradiations.

- From the RT measurements during irradiation it is observed that the self-regulating heater
  on the sample stage needs to be carefully controlled as it will interfere with the measured
  outputs. Thus, the heater should only be used to initialise an initial bias temperature,
  and then it should be switched to manual mode where the power should be constant
  throughout any irradiation events.

- Temporal voltage measurements is better than real-time RT measurements for Cobalt-60
  irradiation. It can be seen that RT measurements is more susceptible to measurement
  errors.

- A full refill of Liquid Helium to the experimental dewar can last for approximately a day
  of irradiation experiments. This raises the requirement for the liquid Helium transfer
  facility to be moved closer to the Theratron for future irradiation experiments.

An immediate future extension for this work is to measure the overall heat capacity of the
absorber. From the measured $\Delta T$ from ISTED and the heat capacity, the absorbed energy can be
calculated, shown in Chapter 5. Also an area worth exploring is the use of different energy and
different energetic particles by using GEANT4 to simulate the detector under different broad
beam environments.
Figure 8.26: Voltage response from $^{60}\text{Co}$ irradiation.

The effect of Co60 irradiation on the Ic of SQUID C30

Figure 8.27: RT response for pre- and post- and different irradiation dosages.
Figure 8.28: Repeats for two different dosages.
Chapter 9

Conclusions

It is known in the literature that macroscopic radiation qualities such as LET does not relate well to radiobiological effectiveness for when the energy of the incident particles are low [28]. This is because it does not capture the stochastic characteristics of particle track structures. For low energy particles such as sub 1 keV electrons, the clustering of ionisation interactions on nanometre scale is a key parameter to the determination of the lesion damage and the survival rate of the cell. On the other hand, nanodosimetric quantities such as ionisation cluster size distributions are active areas of research which aims to provide the needed physical quantities to relate to DNA damages [68, 17].

However, there is also a risk in only using the current existing nanodosimetric quantities as they are all related to ionisation interactions, where all other interactions are neglected. Biological systems are actually very complex and the consideration of only one type of interaction may be an over simplification to the problem. It is seen in many experiments that other interaction events such as DNA molecule excitations and formations of free radicals are also important pathways to serious DNA lesions [26, 101, 19, 130]. Thus, the design of ISTED as a nano-calorimeter fits into the missing gap by providing measurements on the energy distribu-
tion profiles on the interaction events from particles of interest with solid-state.

In Chapter 4, I have shown that the study of nanodosimetric physical quantities are unaffected by the fuzzy boundary caused by quantum to classical mechanics. Furthermore, I have also shown that the extreme high sensitivity and ultra-low noise performance of nanobridge Josephson junction makes it an ideal component for ISTEDs. Nanobridge Josephson junctions fabricated using the dual-bean FIB technique is observed to produce devices with consistently good performances. The magnetic flux noise of nanoSQUID is found as $3 \times 10^{-14} \Phi_0^2$ in the white noise region, which is sufficient for the measurement of sub 100 eV energy absorption distribution for low energetic electron interactions.

9.1 Immediate steps and simple improvements

9.1.1 SQUID design

The performance of a SQUID can be enhanced when coupled with a well-designed pickup loop. For SQUID magnetometers, a common configuration for pickup loops are in the form of square washers. A square washer is a wide superconducting loop that is essentially a single-turn flux transformer. Like any superconducting loop, it will exclude magnetic fields from its film where parts of the diverted fields will be focused into the inner hole. For square washers with loop width that is at least in the same order of magnitude has a self-inductance, $L$, that is proportional to its hole width, $d$. Simple improvements on the SQUID design can be realised with the simulation package 3DMLSI outlined in Chapter 6.
9.2  Future works

9.2.1  Micro-Beam and Nano-beam alignments

In the long term, quantification of the the energy absorption distribution for low energy electrons can be achieved through the use of the micro-beam or nano-beam (when it becomes available) at the Ion Beam Centre at The University of Surrey. However, this goal will involve a lot intermediate steps and would take the effort of another PhD student perhaps in the future.

9.2.2  Biological targets

As the top layer absorber film used in the current design of the detector is made from Carbon, one possible way to reduce the uncertainties in the conversion of measured energy absorption of Carbon to that of biological cellular mediums is to substitute it with biological targets. This will better benefit research in micro- and nanodosimetric model developments for thin-film cell survival protocols with low energy electrons.

9.3  A final note

Nanodosimetry is an important area of research within the grander community of radiation protection and radiotherapy, where improving the understanding of radio-biological effects are an important driver to the fields. Superconducting detectors such as ISTED demonstrated huge potentials in improving the accuracies and precisions in current available nanodosimetric physical quantities. In my views, the unparalleled sensitivity in nanoSQUIDs should be exploited to the fullest extent in the future.
Appendices
Appendix A

Publications and conference presentations

A.0.1 Thesis Publications


A.0.2 Other Publications


A.0.3 Presentations


3. Patel, T., Li, B., Gallop, J., Cox, D., Kirkby, K., Romans, E., Chen, J., Nisbet, A. and Hao, L.,


Appendix B

Source Codes

The codes used in my work on the simulation of ionisation cluster-size distributions are based from the example codes provided by the GEANT4 Collaboration.

Below are source codes to the GEANT4 classes I have used in my application in Chapter 4:

Listing B.1: GEANT4-DNA license

```c
// ********************************************************************
// * License and Disclaimer
// * *
// * The Geant4 software is copyright of the Copyright Holders of *
// * the Geant4 Collaboration. It is provided under the terms and *
// * conditions of the Geant4 Software License, included in the file *
// * LICENSE and available at http://cern.ch/geant4/license. These *
// * include a list of copyright holders. *
// * *
// * Neither the authors of this software system, nor their employing *
```

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Listing B.2: Primary Generator Action Class

#include "PrimaryGeneratorAction.hh"

#include "G4SystemOfUnits.hh"

#include "DetectorConstruction.hh"

#include "G4Event.hh"

#include "G4ParticleTable.hh"

#include "G4ParticleDefinition.hh"

PrimaryGeneratorAction::PrimaryGeneratorAction(DetectorConstruction* detectorConstruction) : G4VUserPrimaryGeneratorAction(),
    fParticleGun(0),

fDetConstruction(detectorConstruction)
{
  G4int n_particle = 1;
  fParticleGun = new G4ParticleGun(n_particle);

  G4ParticleTable* particleTable = G4ParticleTable::GetParticleTable();
  G4String particleName;
  G4ParticleDefinition* particle
    = particleTable->FindParticle(particleName="e-");

  fParticleGun->SetParticleDefinition(particle);
  fParticleGun->SetParticleEnergy(100.*eV);
  fParticleGun->SetParticleMomentumDirection(G4ThreeVector(0.,0.,1.));
  fParticleGun->SetParticlePosition(G4ThreeVector(0.,0.,0.));
}

PrimaryGeneratorAction::~PrimaryGeneratorAction()
{
  delete fParticleGun;
}

void PrimaryGeneratorAction::GeneratePrimaries(G4Event* anEvent)
{
  fParticleGun->GeneratePrimaryVertex(anEvent);
}
Listing B.3: Physics List Class

```cpp
#include "PhysicsList.hh"
#include "G4SystemOfUnits.hh"
#include "G4EmDNAPhysics.hh"

PhysicsList::PhysicsList() : G4VModularPhysicsList()
{
    currentDefaultCut = 1.0*micrometer;
    cutForGamma = currentDefaultCut;
    cutForElectron = currentDefaultCut;
    cutForPositron = currentDefaultCut;
    SetVerboseLevel(1);
    emPhysicsList = new G4EmDNAPhysics();
}

PhysicsList::~PhysicsList()
{
}

void PhysicsList::ConstructParticle()
{
    emPhysicsList->ConstructParticle();
}
```
void PhysicsList::ConstructProcess()
{
    AddTransportation();
    emPhysicsList->ConstructProcess();
}

#include "G4Gamma.hh"
#include "G4Electron.hh"
#include "G4Positron.hh"

void PhysicsList::SetCuts()
{
    G4ProductionCutsTable::GetProductionCutsTable()->
        SetEnergyRange(100*eV, 1*GeV);
    SetCutValue(cutForGamma, "gamma");
    SetCutValue(cutForElectron, "e-");
    SetCutValue(cutForPositron, "e+");
}

void PhysicsList::SetCutForGamma(G4double cut)
{
    cutForGamma = cut;
    SetParticleCuts(cutForGamma, G4Gamma::Gamma());
}
void PhysicsList::SetCutForElectron(G4double cut)
{
    cutForElectron = cut;
    SetParticleCuts(cutForElectron, G4Electron::Electron());
}

void PhysicsList::SetCutForPositron(G4double cut)
{
    cutForPositron = cut;
    SetParticleCuts(cutForPositron, G4Positron::Positron());
}

Listing B.4: Detector Construction Class

#include "EventAction.hh"
#include "RunAction.hh"
#include "Analysis.hh"
#include "G4RunManager.hh"
#include "G4Event.hh"
#include "G4UnitsTable.hh"
#include "Randomize.hh"
#include <iomanip>

EventAction::EventAction()
: G4UserEventAction(),
  Energy(0.),
Length(0.),
IonCount(0.),
EventNum(0.),
IonCountTarget(0.)
()

EventAction::~EventAction()
()

void EventAction::BeginOfEventAction(const G4Event* evt)
{
    Energy = 0.;
    Length = 0.;
    IonCount = 0.0;
    EventNum = 0.0;
    IonCountTarget = 0.0;
}

void EventAction::EndOfEventAction(const G4Event* event)
{
    G4AnalysisManager* analysisManager = G4AnalysisManager::Instance();
    fstream text1;
    text1.open("20170407_100eV_electron.txt",std::fstream::app);
    text1<<IonCount<<"\n";
}
text1.close();
fstream text2;

    text2.open("20170407_100eV\_electron_count.txt",std::fstream::app);
text2<"EventNum"<"\n";
text2.close();
}

Listing B.5: Run Action Class

#include "RunAction.hh"
#include "G4Run.hh"
#include "Analysis.hh"

RunAction::RunAction(DetectorConstruction*)
{
}

RunAction::~RunAction()
{
}

void RunAction::BeginOfRunAction(const G4Run* aRun)
{
    G4cout << "#####\_Create\_analysis\_manager" << "\n" << this << G4endl;
    G4AnalysisManager* analysisManager = G4AnalysisManager::Instance();
    G4cout << "Using\_" << analysisManager->GetType()
    << "\_analysis\_manager" << G4endl;
    analysisManager->SetVerboseLevel(1);
G4String fileName = "20171001_1keV_electron_1_5um";
analysisManager->OpenFile(fileName);
analysisManager->CreateNtuple("dna", "dnaphysics");
analysisManager->CreateNtupleDColumn("x");
analysisManager->CreateNtupleDColumn("y");
analysisManager->CreateNtupleDColumn("z");
analysisManager->CreateNtupleDColumn("totalEnergyDeposit");
analysisManager->CreateNtupleDColumn("uncertaintyTotE");
analysisManager->CreateNtupleDColumn("PreVelocity");
analysisManager->CreateNtupleDColumn("Origin");
analysisManager->CreateNtupleDColumn("particle");
analysisManager->CreateNtupleDColumn("process");
analysisManager->FinishNtuple();
fstream text;
fstream text1;
}

void RunAction::EndOfRunAction(const G4Run* aRun)
{
G4int nofEvents = aRun->GetNumberOfEvent();
if ( nofEvents == 0 ) return;
G4AnalysisManager* analysisManager = G4AnalysisManager::Instance();
analysisManager->Write();
analysisManager->CloseFile();
delete G4AnalysisManager::Instance();
void RunAction::save()
{
    text.open("test\save\in RunAction.txt");
    text<<"Trial";
    text.close();
}

void RunAction::save1()
{
    text1.open("test\save\in RunAction1.txt");
    text1<<"Trial";
    text1.close();
}

Listing B.6: Event Action Class

#include "EventAction.hh"
#include "RunAction.hh"
#include "Analysis.hh"
#include "G4RunManager.hh"
#include "G4Event.hh"
#include "G4UnitsTable.hh"
#include "Randomize.hh"
#include <iomanip>
EventAction::EventAction()
 : G4UserEventAction(),
 Energy(0.),
 Length(0.),
 IonCount(0.),
 EventNum(0.),
 IonCountTarget(0.)
 {}

EventAction::~EventAction()
 {}

void EventAction::BeginOfEventAction(const G4Event* evt)
 {
  Energy = 0.;
  Length = 0.;
  IonCount = 0.0;
  EventNum = 0.0;
  IonCountTarget = 0.0;
 }

void EventAction::EndOfEventAction(const G4Event* event)
 {
  G4AnalysisManager* analysisManager = G4AnalysisManager::Instance();
fstream text1;
text1.open("20170407_100eV_electron.txt", std::fstream::app);
text1<<IonCount<<"\n";
text1.close();
fstream text2;
text2.open("20170407_100eV_electron_count.txt", std::fstream::app);
text2<<EventNum<<"\n";
text2.close();
}

Listing B.7: Stepping Action Class

#include "Analysis.hh"
#include "SteppingAction.hh"
#include "RunAction.hh"
#include "DetectorConstruction.hh"
#include "PrimaryGeneratorAction.hh"
#include "G4SystemOfUnits.hh"
#include "G4SteppingManager.hh"
#include "G4Electron.hh"
#include "G4Proton.hh"
#include "G4Gamma.hh"
#include "G4Alpha.hh"
#include "G4DNAGenericIonsManager.hh"
#include "EventAction.hh"
#include "G4Step.hh"
#include <fstream>
using namespace std;

SteppingAction::SteppingAction(EventAction* eventAction,
DetectorConstruction* detectorConstruction)
  : G4UserSteppingAction(),
    fEventAction(eventAction),
    fDetConstruction(detectorConstruction)
{}

SteppingAction::~SteppingAction()
{}

void SteppingAction::UserSteppingAction(const G4Step* step)
{
  flagParticle=-1.;
  flagProcess =-1.;
  if (step->GetTrack()->GetDynamicParticle()->GetDefinition()->
      GetParticleName() == "e-")
    flagParticle = 1;
  if (step->GetTrack()->GetDynamicParticle()->GetDefinition()->
      GetParticleName() == "proton")
    flagParticle = 2;
  if (step->GetTrack()->GetDynamicParticle()->GetDefinition()->
      GetParticleName() == "hydrogen")

flagParticle = 3;

if (step->GetTrack()->GetDynamicParticle()->GetDefinition() ->
GetParticleName() == "alpha")
flagParticle = 4;

if (step->GetTrack()->GetDynamicParticle()->GetDefinition() ->
GetParticleName() == "alpha+")
flagParticle = 5;

if (step->GetTrack()->GetDynamicParticle()->GetDefinition() ->
GetParticleName() == "helium")
flagParticle = 6;

if (step->GetPostStepPoint()->GetProcessDefinedStep()->
GetProcessName() == "e-_G4DNAElastic")
flagProcess =11;

if (step->GetPostStepPoint()->GetProcessDefinedStep()->
GetProcessName() == "e-_G4DNAExcitation")
flagProcess =12;

if (step->GetPostStepPoint()->GetProcessDefinedStep()->
GetProcessName() == "e-_G4DNAIonisation")
flagProcess =13;

if (step->GetPostStepPoint()->GetProcessDefinedStep()->
GetProcessName() == "e-_G4DNAAttachment")
flagProcess =14;

if (step->GetPostStepPoint()->GetProcessDefinedStep()->
GetProcessName() == "e-_G4DNAVibExcitation")
flagProcess =15;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="proton_G4DNAExcitation")
    flagProcess =17;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="proton_G4DNAIonisation")
    flagProcess =18;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="proton_G4DNAChargeDecrease")
    flagProcess =19;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="hydrogen_G4DNAExcitation")
    flagProcess =20;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="hydrogen_G4DNAIonisation")
    flagProcess =21;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="hydrogen_G4DNAChargeIncrease")
    flagProcess =22;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="alpha_G4DNAExcitation")
    flagProcess =23;
if (step->GetPostStepPoint()->GetProcessDefinedStep()->GetProcessName()=="alpha_G4DNAIonisation")
    flagProcess =24;
GetProcessName ()=="alpha_G4DNAChargeDecrease")
    flagProcess =25;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="alpha+_G4DNAExcitation")
    flagProcess =26;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="alpha+_G4DNAIonisation")
    flagProcess =27;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="alpha+_G4DNAChargeDecrease")
    flagProcess =28;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="alpha+_G4DNAChargeIncrease")
    flagProcess =29;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="helium_G4DNAExcitation")
    flagProcess =30;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="helium_G4DNAIonisation")
    flagProcess =31;
if (step->GetPostStepPoint ()->GetProcessDefinedStep ()->
GetProcessName ()=="helium_G4DNAChargeIncrease")
    flagProcess =32;
if ( (step->GetPostStepPoint ()->GetProcessDefinedStep()->
GetProcessName ()!="Transportation"))
if (flagProcess != 0) {
    xp = step->GetPostStepPoint()->GetPosition().x() / nanometer;
    yp = step->GetPostStepPoint()->GetPosition().y() / nanometer;
    zp = step->GetPostStepPoint()->GetPosition().z() / nanometer;
    TargetRadius = 10*nm/2;
    TargetZ = 10*nm;
    r = sqrt((xp*xp) + (yp*yp));
    if (r <= TargetRadius && zp <= TargetZ && zp >= 0) {
        hbar = 1.0545717260 / pow(10.0, 34.0);
        totEnergy = step->GetTotalEnergyDeposit();
        velocity = step->GetPreStepPoint()->GetVelocity() / (m/s);
        uncertainty2 = (hbar * velocity * pow(10.0, 9.0)) / (totEnergy / joule);
        G4AnalysisManager* analysisManager = G4AnalysisManager::Instance();
        analysisManager->FillNtupleDColumn(0, xp);
        analysisManager->FillNtupleDColumn(1, yp);
        analysisManager->FillNtupleDColumn(2, zp);
        analysisManager->FillNtupleDColumn(3, totEnergy / eV);
        analysisManager->FillNtupleDColumn(4, uncertainty2);
        analysisManager->FillNtupleDColumn(5, velocity);
        analysisManager->FillNtupleDColumn(6, 0);
    }
}
analysisManager->FillNtupleDColumn(7, flagParticle);

analysisManager->FillNtupleDColumn(8, flagProcess);

analysisManager->AddNtupleRow();

fEventAction->AddCount();


[23] D. Charlton, D. Goodhead, W. Wilson, and H. Paretzke. The deposition of energy in small cylindrical targets by high let radiations. Radiation Protection Dosimetry, 13(1-4):123–125,
1985.


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