

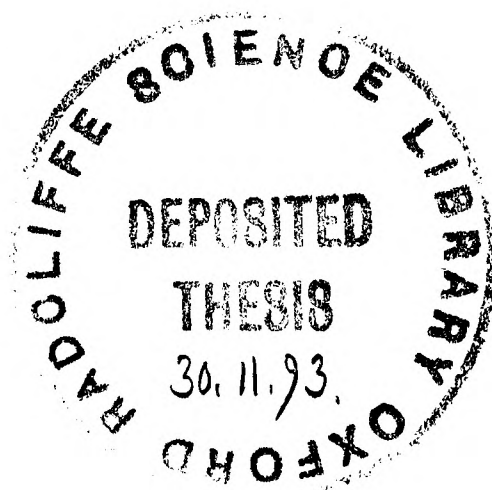
**NON-EQUILIBRIUM SUPERCONDUCTIVITY IN
NIOBIUM
AND ITS APPLICATION TO PARTICLE DETECTION**

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ABSTRACT

The propagation of non-equilibrium phonon and quasiparticle signals in bulk single crystal superconducting niobium has been investigated using superconducting tunnel junctions. Excitations generated by pulsed laser light and particle interactions were studied in 2–12 g crystals. At temperatures below 2 K the lifetimes of non-equilibrium phonons were found to be very much shorter than the theoretical bulk anharmonic decay times. Quasiparticle propagation was observed limited by impurity scattering. The proximity effect in Al thin films on Nb single crystal surface was studied. Particle interactions were detected in Nb with both position and energy resolution. The possibilities for using a detector based on superconducting single crystal niobium for a dark matter search are discussed.

For Serena

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CONTENTS

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1. THESIS INTRODUCTION

After a year's research, one realises that it could have been done in a week.

Sir William Henry Bragg

1.1 INTRODUCTION

In particle physics it is becoming increasingly difficult to probe further into the structure of matter. Accelerator experiments are used to create particles at high energies. However, a number of fundamental problems in cosmology, neutrino and weak interaction physics, that do not require high energy accelerators, have not been resolved for lack of sensitive particle detectors. This work on the propagation of non-equilibrium signals in superconducting Niobium was catalysed by one such problem.

The search for weakly interacting massive particles (WIMPs) ¹ by observing their scattering in underground detectors will require a range of target materials. Nb is a good candidate given it is mono-isotopic with a large neutron content ($N=52$) and non-zero nuclear spin ($9/2$). This makes it sensitive both to the coherent scattering of WIMPs through vector interactions and to axial couplings.

¹These particles have been proposed as a solution to the “missing” mass or dark matter problem of the Universe. This is covered in Chapter 2.

The purpose of this work was to study the propagation of phonons and quasiparticles produced in single crystal superconducting Nb by laser and particle interactions. Superconducting Tunnel Junctions (STJs) and Series Arrays of Al Superconducting Tunnel Junctions (Al SASTJs) fabricated on the surface of Nb crystals were used to detect these signals.

1.2. GLOBAL DEVELOPMENT OF LOW TEMPERATURE DETECTORS

This work should be viewed in the context of the wider development of particle detectors based on low temperature (less than 4 K) techniques. These new detectors can be divided into two broad categories which are based either on the detection of thermal phonon distributions (calorimeters) or more complex non-equilibrium signals. The dividing line is, however, becoming increasingly blurred.

Using calorimetric techniques McCammon and Moseley [1] have fabricated small mass bolometers with resolutions close to their theoretical limit. In a 10^{-5} g Si HgCdTe absorber ² they have a baseline energy fluctuation of 4.5 eV for 6 keV x-rays.

In much more massive calorimeters the Sadoulet group [2] at Berkeley now have a FWHM resolution of < 2 keV for 60 keV γ -rays in a 60 g Ge absorber at 30 mK.³ This detector also demonstrates another interesting development in cryogenic detectors. The simultaneous read-out of both the thermal and ionisation signals ⁴ from the Ge absorber allows the separation of neutron and electron recoils ⁵.

The Garshing-TUM Group are investigating superconducting absorbers (V and Mo) using superconducting transition edge bolometers [4]. They currently have a resolution of 1.2% FWHM for 5.5 MeV α -particles. Their work will be interesting as it will provide more information about the behaviour of non-thermal and thermal signals in superconducting absorbers similar to Nb.

²The temperature rise in the absorber was detected using an ion implanted Si thermistor.

³The temperature rise in the absorber was detected using neutron transmutation doped (NTD) Ge.

⁴See also Spooner *et al.*, UK Dark Matter Collaboration [3]

⁵This is one of the requirements of the next generation of WIMP detectors. (See Chapter 2)

STJs have been widely applied to the detection of particle interactions in the junctions themselves and (more applicable to this work) as detectors of the non-equilibrium signals in large mass (few g) detectors. Work at this laboratory by Goldie on 2 g InSb absorbers using SASTJs has shown resolution FWHM of less than 500 eV on 22 keV X-rays at 70 mK [5].

Transition edge sensors (TES) made from superconducting Ti and W films on Si substrates have been studied extensively by the Cabrera group at Stanford [6, 7]. They are being operated in a mode that makes them sensitive to non-equilibrium phonon signals at surface energy densities as low as $0.5 \text{ meV } \mu\text{m}^{-2}$ which permits the detection of 6 keV X-rays across 500 μm thick Si.

1.3 THESIS STRUCTURE

The demands of a low temperature Nb based detector for dark matter are exacting. If such a device is to be realised a detailed study had to be made of the behaviour of phonons and quasiparticles in bulk Nb single crystals. In particular, the detector would be required to differentiate between nuclear and electron recoils. We have not yet achieved this goal. However, considering this point has raised a number of interesting questions about the way in which interactions couple to the crystal lattice and superconducting state and how this will influence the subsequent spread of the non-equilibrium state throughout the crystal. We have gone some way answering these questions in this work.

In Chapter 2 we discuss some of the theories concerning dark matter and the requirements for a WIMP detector.

Chapter 3 provides a theoretical analysis of the superconducting state in Nb. It also details the response of STJs to non-equilibrium and quasi-thermal signals. It was necessary to develop an understanding of the way in which STJs work so that the correct interpretation of the results from the Nb experiments could be made.

Chapter 4 describes the experimental techniques used to fabricate STJs on the surface of the Nb crystals. The cryogenic systems, laser system and measuring electronics are also described.

Chapter 5 covers a number of the experiments performed to investigate the transport properties of phonons and quasiparticles in Nb single crystals at low temperatures. The results represent only a small – but successful – fraction of the work carried out. Comparisons are made with the theory developed in Chapter 3.

Chapter 6 presents work carried out to study the surface of the Nb crystals and to look at its modification by a thin over-layer of Al. The surface physics of Nb is extremely complex and has posed a number of barriers to the successful study of the bulk properties of Nb. The techniques developed may ultimately assist the detection of non-equilibrium quasiparticle signals through the mechanism of quasiparticle trapping.

Chapter 7 presents some results on the detection of α -particle and γ -radiation and in the light of these results discusses the prospect of differentiating between nuclear and electron recoil events.

Chapter 8 provides a summary of the results presented.

2. DARK MATTER DETECTION

Don't let me catch anyone talking about the Universe in my department.

Lord Rutherford

2.1. INTRODUCTION

There is a considerable weight of evidence that we have yet to discover the major component of matter in the universe. This “missing mass” may take the form of a new particle, as yet unobserved, that permeates all space, including our own galaxy. The motivation for this research on superconducting Nb is to look at the feasibility of constructing a detector for these particles. It may be possible to detect these particles by observing their occasional recoil from a nucleus in a Nb crystal lattice cooled to low temperatures. An interaction will create a non-equilibrium signal (of phonons and quasiparticles) that can be detected at the crystal surface (see Fig. 2.1.1). In this Chapter we will briefly discuss ⁶ the experimental and theoretical background to the problem of the “missing mass” of the Universe. We will also consider the requirements for a low temperature detector to be used in a search for a particular category of particles that may be part of this missing mass.

⁶The missing mass problem and the hunt for candidates to explain it has been reviewed extensively. See for instance Smith and Lewin [8] and Primack, Seckel and Sadoulet [9].

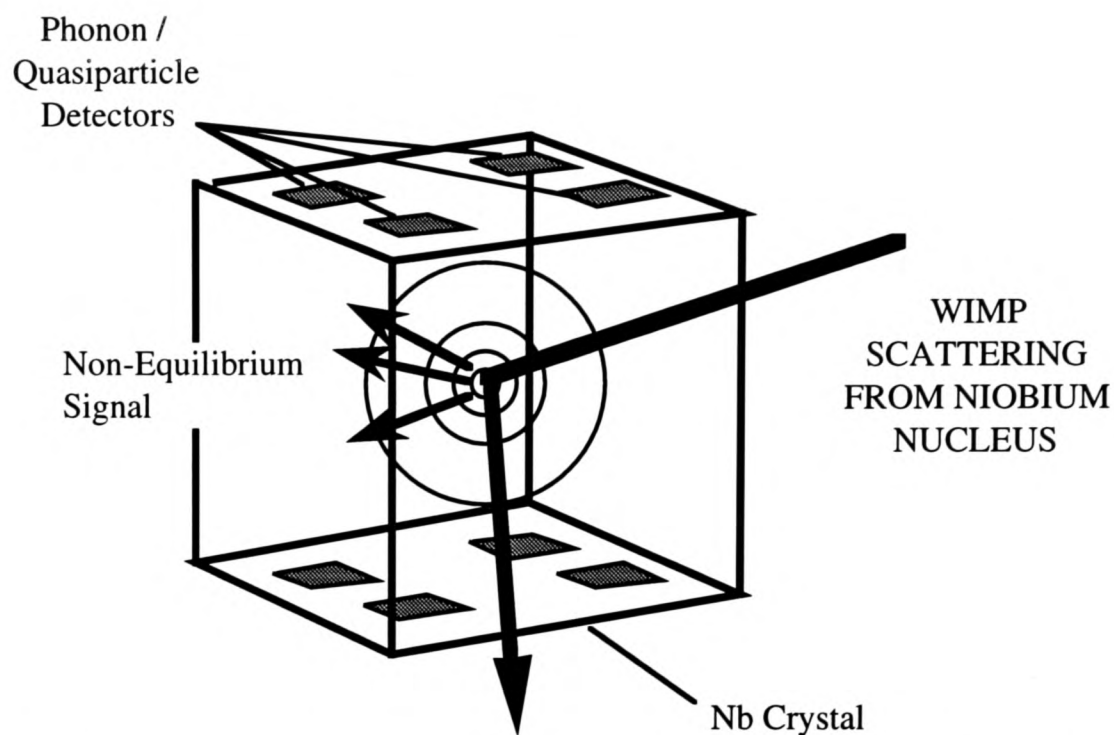


Figure 2.1.1 Schematic of the recoil of a particle from a nucleus within a Nb crystal lattice at low temperatures. The interaction generates a non-equilibrium phonon and quasiparticle signal which can be detected at the surface of the crystal (After Cabrera *et al.* [10]).

2.2. THE EVIDENCE FOR DARK MATTER

2.2.1. $\Omega = 1$? The Mass Density of the Universe

The gravitational behaviour of the rotation of galaxies and clusters of galaxies appear to violate Newton's Law of Gravitation, if it assumed that the mass in a galaxy is distributed in proportion to its luminosity. In the spherically symmetric approximation, the velocity of rotation of the matter in the galaxy would be expected to vary according to the relationship

$$v = \left(\frac{GM}{r} \right)^{\frac{1}{2}}, \quad [2.2.1]$$

where r is the radius from the galactic centre, and M is the mass enclosed in the radius, r . At the edge of a galaxy, where the luminosity falls to a few percent of that at the centre, and so M becomes roughly independent of r , we would expect the velocity, $v \sim r^{-1/2}$. A number of observations (the first by Zwicky in 1933) have shown that the velocity is, in

fact, approximately a constant. In order to explain these observations it has been suggested that a halo of non-luminous (or dark) matter also permeates the galaxies.

If we now consider the gravitational behaviour of the Universe as a whole, it is strongly argued that the total matter in the universe is just sufficient for gravitational forces⁷ to close it. If we denote the mean density of the Universe (for closure) as $\Omega = 1$, then it is argued that if $\Omega > 1$ the Universe would be collapsing under gravitational attraction driving $\Omega \rightarrow \infty$. If $\Omega < 1$ then the Universe would be rapidly expanded, driving $\Omega \rightarrow 0$. If we are to assume that current (large scale) observations which show $\Omega \sim 1$ are not due to the chance that we exist at an era when this just happens to be the case, then $\Omega \equiv 1$. A mass density, $\Omega = 1$ is also predicted by Inflation Theory, which describes the early expansion of the Universe, after the Big Bang. This theory helps to resolve problems concerning flatness and the event horizon and is highly favoured.

Astronomical measurements of the mass density of the Universe yield a range of values which appear to get closer to $\Omega = 1$ at larger distance scales (see Table 2.2.1).

	Scale ($\times h^{-1}$ Mpc)	Ω
Luminous parts of galaxies	~ 0.02	0.01
Haloed of galaxies and groups of galaxies	$\sim 0.1-1$	0.02-0.2
Cosmic virial theorem	~ 3	~ 0.2
Virgo infall	~ 10	~ 0.2
Large-scale infall	~ 30	$\sim 0.2-1$
Cosmological tests	3000	0.1-2
Cosmic Inflation		1

Table 2.1.1. Ω , as a function of distance scale. (After [9]). h is a parameter ~ 1 that reflects the uncertainty in the Hubble Constant.

Luminosity measurements of the centres of spiral galaxies indicate that the density of luminous (baryonic) matter is $\Omega_B \leq 0.01$, less than 1% of the density required to close

⁷On cosmology scales gravity becomes the dominant force

the Universe ⁸. If we include the galaxy dark matter halos, mentioned in the first paragraph, this increases the total mass density to $\Omega \sim 0.03\text{--}0.10$. Observations of the gravitational behaviour of galactic clusters seems to indicate additional halo material spreading out on much larger scales which increases Ω still further to $0.1\text{--}0.3$. The independent motions of galaxies ⁹ due to local gravitational fields also provides evidence for $\Omega = 0.2\text{--}0.7$.

Limits on the total amount of baryonic matter in the Universe can be placed by comparing the observed abundances of the light elements ¹⁰ with those predicted by Primordial Nucleosynthesis. The calculations of this model depend on the ratio, η , for the baryon to photon ratio in the Universe. A good match to the observed abundances is found for

$$3 \times 10^{-10} < \eta < 5 \times 10^{-10}.$$

[2.2.2]

The photon number is given by the cosmic microwave background (temperature, 2.71 K) and this constrains the baryon density, $0.01 < \Omega_B < 0.12$. If $\Omega = 1$ then a significant fraction of the Universe is non-baryonic. This situation is summarised in Fig. 2.2.1.

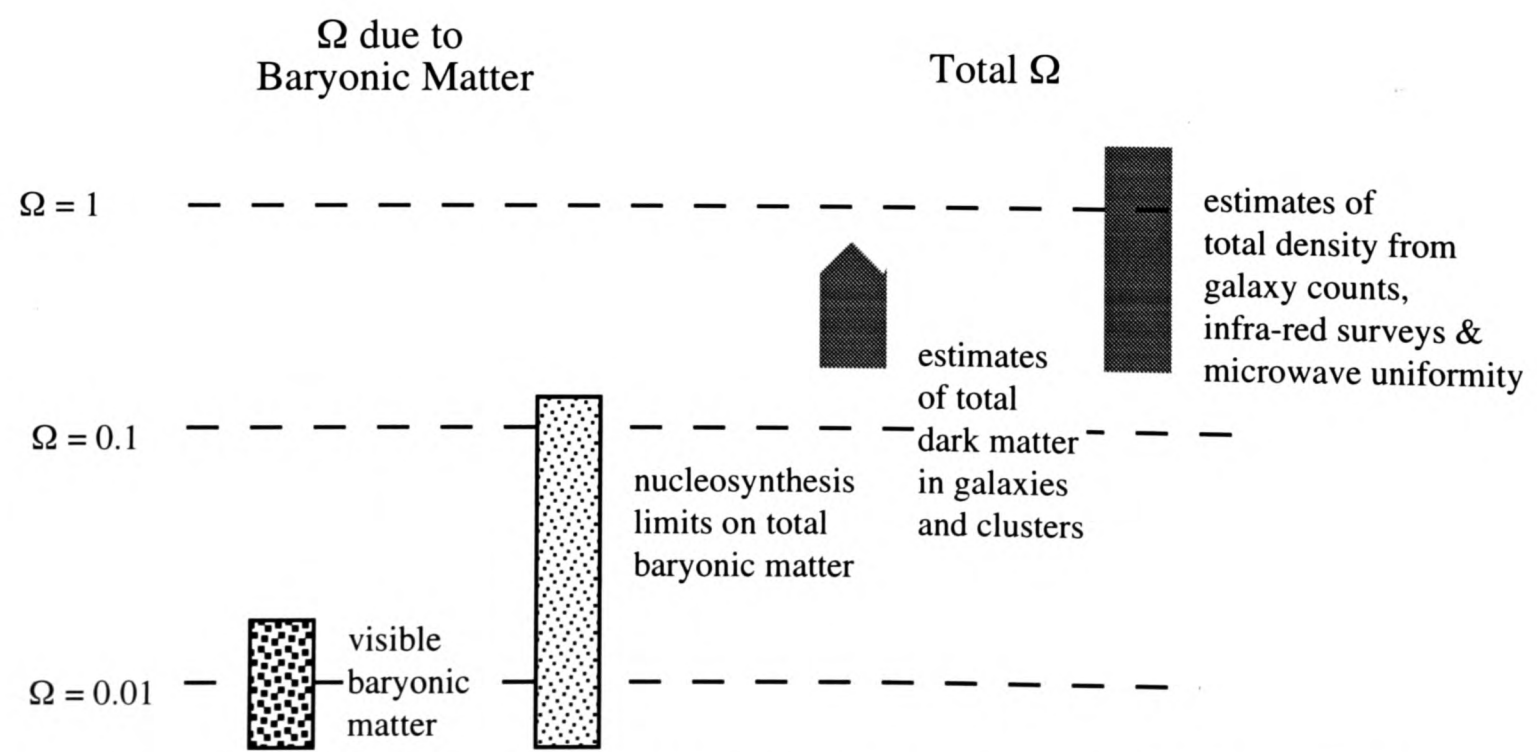


Figure 2.1.1. Estimates of total density parameter Ω and baryonic component (after Smith and Lewin [8]).

⁸We will use Ω_B to denote the mass density of baryonic matter.
⁹Superimposed on the general expansion of the Universe.
¹⁰D, ³He, ⁴He and ⁷Li

2.2.2. Weakly Interacting Massive Particles

The arguments in the previous section indicate that a significant fraction of dark matter is non-baryonic. Candidates currently proposed for this matter ¹¹ include one flavour of the light neutrinos, axions and the lightest spin-half neutral supersymmetric particle (LSP) [12]. These particles are divided into two categories, hot and cold dark matter, so called to indicate their velocities (relativistic and non-relativistic, respectively) at the time they dropped out of thermal equilibrium with the rest of the matter in the early expansion of the Universe. The light neutrino is an example of hot dark matter (HDM) and would have a mass in the range 20–60 eV for the relic density to close the Universe ($\Omega = 1$). Difficulties have been encountered in the failure of HDM to explain the formation of small scale structure (galaxy formation) in the Universe. LSPs (as well as heavy Dirac neutrinos, $m_{\nu D} > 1/2 m_Z$) are examples of cold dark matter (CDM), and are expected to have a mass in the range 1–10³ GeV [13, 14]. Such a particle could be produced in sufficient quantity in the early universe to provide a dominant non-baryonic component of the matter density and would subsequently cluster in association with stellar matter to form galactic halos. Estimates have been made of the likely cross-section of these particles on baryonic matter and are found to be less than or the order of the weak interaction. They have thus been labelled Weakly Interacting Massive Particles (WIMPs). Our investigations into Nb are aimed at the construction of a detector to observe (or exclude) this category of particles.

¹¹Particles that have been proposed and excluded (to varying extents) include the electron neutrino (mass constrained by Super Nova 1987A observations), the second lightest neutrino (mass possibly constrained by consideration of mass mixing with electron neutrino explaining observations in solar neutrino experiments), massive Dirac and Majorana neutrinos with masses less than half the Z mass (results from LEP Z-decay width), sneutrinos (LEP results), Cosmions (eliminated by existing dark matter search results) and magnetic monopoles. The latter remain non-favoured candidates for cold dark matter, however, the search for magnetic monopoles in cosmic rays (see [11] and references therein) has placed stringent limits on their existence.

2.3. REQUIREMENTS FOR THE DETECTION OF WIMPS

2.3.1. Event Rates and Kinematics

A WIMP is expected to interact with the nucleus of one of the atoms in the detector. For an elastic scattering event (non-relativistic) the recoil energy will be

$$E_R = m_W v^2 \left(\frac{m_T m_W}{(m_T + m_W)^2} \right) (1 - \cos \theta), \quad [2.3.1]$$

where m_T and m_W are the masses of the target nucleus and the WIMP, v is the initial velocity, and θ is the scattering angle, of the WIMP. The velocity of the WIMPs in the galaxy ranges between 0 to a cut-off¹² of $\sim 600 \text{ km s}^{-1}$ following a Maxwell-Boltzmann distribution with an r.m.s. velocity $\sim 300 \text{ km s}^{-1}$ [15, 16]. The additional effect of the earth's motion in the galactic frame will be considered in the next section. For a WIMP mass of 20 (100) GeV this gives an average recoil energy (integrating over the WIMP velocity distribution and assuming isotropic scattering from a Nb nucleus) of 5.2 (43) keV.

The cross-section for WIMP interactions is dependent on the type of particle considered. Goodman and Witten [16] have calculated the scattering rate in the non-relativistic limit for any particle that scatters off of nuclei by Z exchange. Their results apply both to massive Dirac neutrinos and to supersymmetric scalar neutrinos. They find that a cross section for scattering of a neutrino of mass m_ν off a nucleus of mass m_T with Z protons and N neutrons is

$$\sigma_{s\nu_D} = \frac{m_\nu^2 m_T^2}{(m_\nu + m_T)^2} \frac{G_F^2 Y^2}{2\pi} [Z(1 - 4 \sin^2 \theta_w) - N]^2 \quad [2.3.2]$$

where G_F is the Fermi constant and Y is the weak hypercharge ($= 1$). Note that in the regime of large de Broglie wavelength for the transfer momentum, the scattering is coherent, and the cross-section is given by the scattering off a single nucleon times the

¹²Determined by the escape velocity from the galaxy.

factor in the square brackets (to the power 2). This factor¹³ is $\sim N^2$ and would favour targets nuclei with a large number of neutrons (see Fig. 2.3.1). For large mass WIMPs there will be a loss of coherence for large recoil energies which will suppress this factor. This has been considered by Ellis and Flores [17] and Engel and Vogel [18] and will become significant for WIMP masses ≥ 50 GeV.

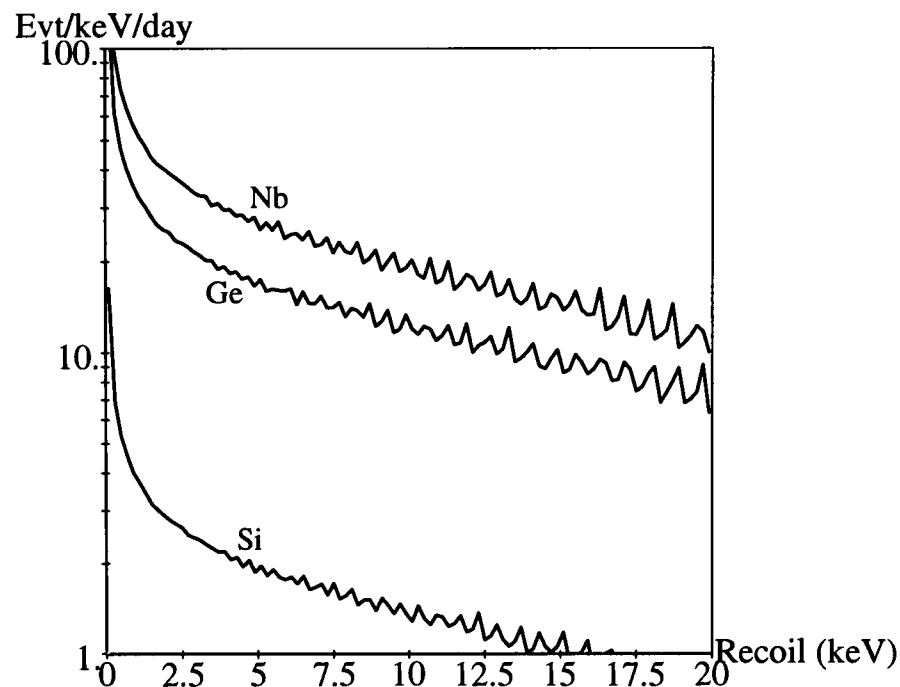


Figure 2.3.1 Calculated recoil spectrum (Events $\text{keV}^{-1} \text{kg}^{-1} \text{Day}^{-1}$) for 50 GeV WIMPs incident on 3 different absorber materials (Si, Ge and Nb). The model assumes cross-section characteristic of weak coherent scattering (Eq. 2.3.2) of WIMPs with a Maxwell-Boltzmann velocity distribution ($v_{\text{RMS}} \approx 300 \text{ km s}^{-1}$ and a cut off at 600 km s^{-1}). No account is taken of the loss of coherence for high momentum transfers during nuclear recoil. The fluctuations in the tails of the distributions are caused by low statistics in the Monte Carlo simulation.

Massive Majorana neutrinos scatter only through spin-dependent interactions. The cross-section for scattering off nucleons

$$\sigma_{sv_M} = \frac{m_v^2 m_T^2}{(m_v + m_T)^2} \frac{G_F^2}{8\pi} \lambda^2 J(J+1) \quad [2.3.3]$$

is much smaller than that for massive Dirac neutrinos, and similar to that for LSPs, which also have axial-vector couplings. λ is a nucleus dependent factor and J is the magnitude of the nuclear spin. For the single particle shell model

¹³Since $\sin^2 \theta_W$ is ~ 0.23 and so the proton (Z) contribution is suppressed.

$$\lambda \sim \pm \frac{1}{2L+1} \quad [2.3.4]$$

for a nucleon in an orbital L and $J = L \pm 1/2$. The values of $\lambda^2 J(J+1)$ have been tabulated for the single particle shell model and also for the odd group nuclear model¹⁴ in Ellis and Flores [17] and Engel and Vogel [18]. Engel *et al.* [19] have considered the case of (neutralino) scattering from Nb specifically, using a model which considers 3 valence protons and 2 valence neutrons. They conclude that in this isolated instance the predictions of the odd group model are worse than those of the single particle shell model.¹⁵

For an LSP the coupling is dominated by the axial component. The cross section (at low energies) is given by Eq. 2.3.3 where the coupling is replaced by a term summed over the quarks charges, q_f and the mass of the scalar particle, m_f , exchanged in the scattering,

$$\sigma_{sLSP} = \frac{m_v^2 m_T^2}{(m_v + m_T)^2} \frac{4q_f^4}{\pi m_f^4} \lambda^2 J(J+1) \quad [2.3.5]$$

The density of WIMPs¹⁶ at the earth is expected to be $\sim 0.3 \text{ GeV cm}^{-3}$ which is about a third of that at the centre of the Milky Way [9]. Ellis and Flores have calculated that the expected total scattering rate for neutralinos (LSPs), of mass 25 and 75 GeV, on ^{93}Nb , is in the approximate range $0.002\text{--}0.1 \text{ events kg}^{-1} \text{ day}^{-1}$. The rate is very dependent of the mass of the neutralino and the mass and the nature of the mediating particle.

If further WIMP searches (using earth based recoil detectors) are undertaken it will be necessary to use a range of absorber materials in order to maximise the region of WIMP “mass versus coupling” space¹⁷ that can be checked. In general, the recoil energy of the nucleus will be a maximum for $m_W \sim m_T$, the mass of the target nucleus. Searches for WIMPs that interact via axial coupling will require nuclei with non-zero nuclear spin.

¹⁴This model is normally a better approximation for nuclei away from shell closure. It uses the assumption that the odd group of nucleons contributes all the nuclear angular momentum, while still assuming that the even group of nucleons has a vanishingly small angular momentum.

¹⁵This paper also contains calculations of the nuclear form factors applicable in the analysis of high momentum exchanges.

¹⁶Assuming they are the majority of dark matter in our galaxy.

¹⁷An example of a mass versus coupling diagram is shown in Fig. 2.3.2

Nb is present as a single isotope ¹⁸ with a spin of 9/2. The nuclear spin in Nb derives from an unpaired proton in contrast to that for the Ge and Si targets of existing WIMP recoil experiments which are even-Z with some of their isotopes odd-N. If a WIMP event spectrum is observed in recoil experiments a Nb target would provide a test of the prediction that the coupling to the unpaired nucleon is around 3 times higher for protons versus neutrons.

2.3.2. Background Signals

It was shown in the last section that the expected interaction rates for LSP WIMPs will be extremely low ($< 0.1 \text{ event kg}^{-1} \text{ day}^{-1}$). In order to see these signals it will be necessary to reduce the effective background level to this order.

At the Earth's surface, the flux of cosmic muons is $\sim 2 \times 10^2 \text{ m}^{-2}\text{s}^{-1}$. By locating a detector below ground the muon flux can be significantly reduced. The rate of muon-related events in a detector can be reduced to $< 0.1 \text{ events kg}^{-1} \text{ day}^{-1}$ for depths $\geq 2000 \text{ m.w.e.}$ ¹⁹ Events that pass directly through the detector can be easily identified by a veto. The signals arising from the interaction of cosmic rays with materials outside the detector which lead to a delayed nuclear decay and emission of either γ -ray or neutron radiation are more difficult to eliminate and require care in the design of the passive shielding around the detector.

Radioactive contamination by natural elements will represent a major source of background events coming from the detector itself, the equipment surrounding it and the rock surrounding the underground laboratory. The lowest backgrounds underground sites have measured an unshielded photon interaction rate below 100 keV in 500 g Ge detector (which is comparable to the size of a low temperature detector for dark matter searches) of $\sim 10^6 \text{ kg}^{-1} \text{ day}^{-1}$ [8]. This can be attenuated using shielding materials but the radio purity of these materials then becomes important. High purity refining techniques allow the production of some metals (for use in shielding and cryostat construction) with

¹⁸This will make subsequent analysis of recoil spectra more readily interpretable.

¹⁹The overburden of rock measured in terms of its metres of water equivalent.

extremely low levels of natural radioactivity. Care has to be taken not to induce additional activity from comic ray bombardment and so equipment and detectors must be stored underground. Shielding (boron-loaded polythene, for example) also has to be introduced to reduce the high energy and thermal neutrons fluxes. Neutron events are particularly problematic since they mimic the nuclear recoil events of dark matter scattering. This will be discussed in a later paragraph.

A measurement was made of the radio purity of the high purity Nb samples we have used in our work to date and is shown in Table 2.3.1 [20].

Element	Concentration (ppm)
K	< 0.037
Rb	< 0.0057
Lu	< 0.0010
Th	< 0.00028
U	< 0.00029

Table 2.3.1 Analysis of high purity Nb for levels of elements which are a source of radioactive background.

In addition to the passive shielding techniques discussed above it may also be possible to reduce the effective background event level using other techniques. It has been shown that low temperature Ge and Si detectors, which simultaneously measure the ionisation and phonon yield from an interaction, can distinguish between γ -ray and neutron recoil events ²⁰. We are investigating the dynamics of interactions in Nb in order to establish if such discrimination is possible by either measuring the relative phonon and quasiparticle yields or the spectrum of non-equilibrium phonons . Some initial results for Nb are reported on α -particle and γ -ray events (in Chapter 7) and also on the significant variation in pulse shapes for different non-equilibrium phonon distributions (in Chapter 5).

²⁰See the Proceedings of the IVth International Workshop on Low Temperature Detectors for Neutrinos and Dark Matter, Oxford, UK [21].

WIMPs might give unique signals in detectors that would distinguish them from the background. It has been calculated that the variation of the velocity of the earth through the galaxy, due to its motion around the Sun, will impose a small annual perturbation on the average recoil energy and event rate of a few percent[9]. An statistical analysis based on this effect will, however, remain difficult given the small number of expected events.

The variation of observed WIMP interaction rates for different target materials (different nuclear masses and/or spins) may also provide evidence for genuine observation of recoils. Many of the techniques for detection of phonon signals reported in this work could be applied to other materials although, as has become apparent during our investigation of Nb, the nature of the transport of non-equilibrium signals in single crystals can be complex and unexpected.

2.3.3. Existing Dark Matter Limits based on Recoil Experiments

The present limits on WIMP dark matter, from the LBL/UCSB Oroville experiments, are shown in Fig. 2.3.3. The solid lines represent the 3σ exclusion limits on the mass-coupling of WIMPs, excluding those parameters to the right and above the lines. The left-hand vertical parts of the lines are limited by the detector thresholds which determine the minimum recoil energy observable²¹. The limits running upwards and to the right are constrained by the background event rates in the detectors. Since the mass density of WIMPs is fixed, as the mass of the WIMPs increases their number density falls and so does the probability of their interacting. The best current background levels in these detectors are $\sim 1 \text{ event keV}^{-1} \text{ kg}^{-1} \text{ day}^{-1}$ [23]. Comparable results for dark matter limits, using Ge detectors, have also been reported by other groups [24, 25].

²¹ The results were taken with conventional Si and Ge semiconductor ionisation detectors. The best 1 kg detectors have thresholds for detecting nuclear recoil at energies ~ 2 and 6 keV , respectively. It should be remembered that (for recoils $\sim 4 \text{ keV}$) only 25% of the nuclear recoil energy appears as ionisation when compared to the ionisation yield from an electron recoil of the same energy.[22].

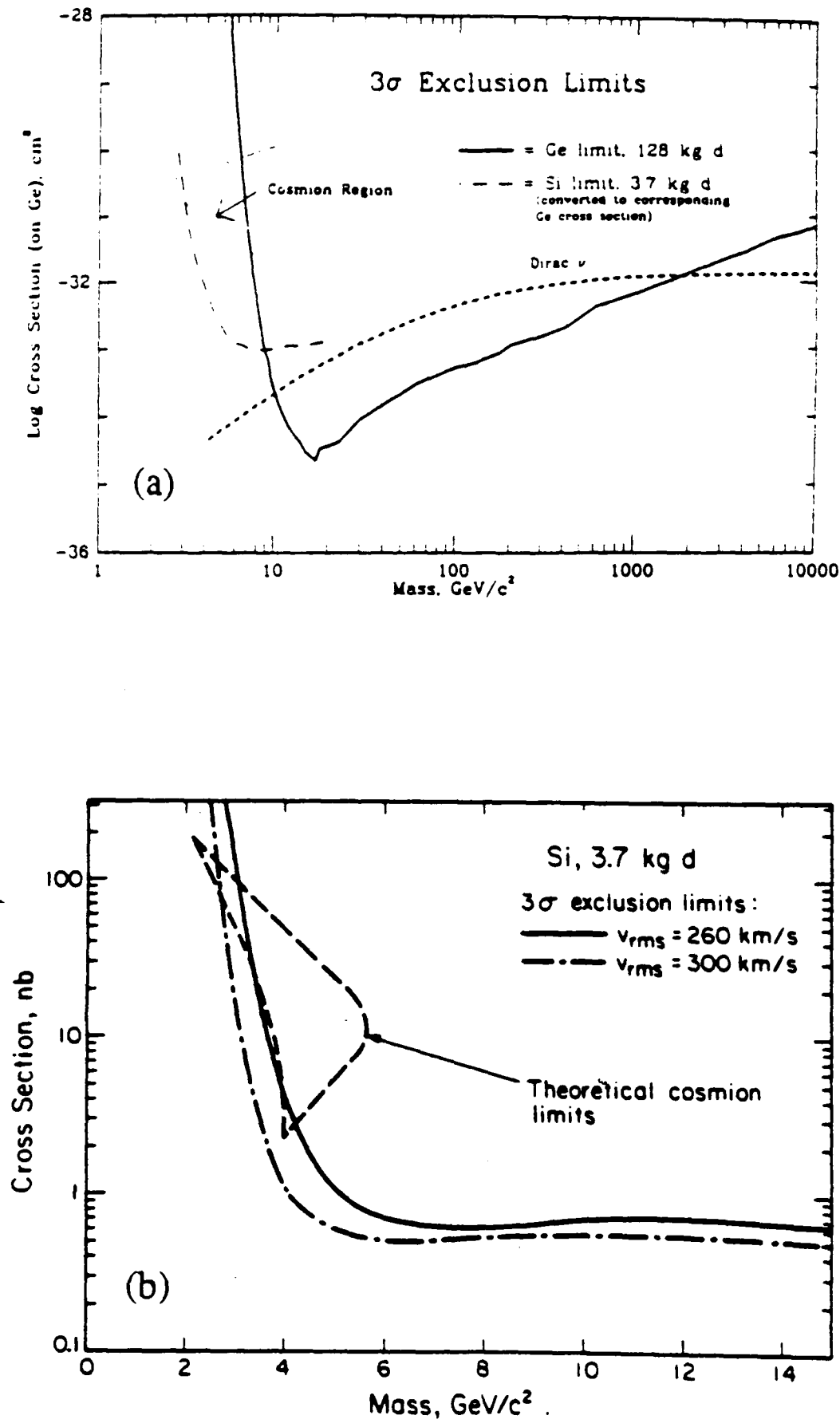


Figure 2.3.2 Limits on mass and cross section of dark matter particles obtained with (i) Ge ionisation and (ii) Si ionisation detectors.[26]

3. THEORETICAL TREATMENT OF
SUPERCONDUCTING NIOBIUM AND
SUPERCONDUCTING TUNNEL JUNCTIONS

The purpose of models is not to fit the data but to sharpen the questions.

Samuel Karlin

3.1. INTRODUCTION

In this chapter we will discuss the theoretical background of non-equilibrium effects in superconductors. Principally, this will involve the calculation of the characteristic rates for exchange, intra- and inter-, the phonon and quasiparticle systems in Nb. In Chapter 4 we will use these rates to interpret our experimental results. Previous experimental studies of other bulk superconductors are listed in Table 3.1.1.

Bulk Superconductor	
Narayanamurti et al. [27]	Pb
Pannetier and Maneval [28-30]	Sn
Goldie [31]	In

Table 3.1.1. Previous experimental investigations of bulk superconducting single crystals.

This work reports results about Nb that differ significantly from those obtained from the studies of Pb, Sn and In. This divergence can be attributed mainly to the

differences in the characteristic times, governing the behaviour of the phonons and quasiparticles.

In the course of the experimental investigations in this work it was surprising to discover that the effective propagation times of the phonon and quasiparticles are reversed in Nb.²² While the velocity of phonons in Nb is 100 times lower than that of the quasiparticles²³, the latter have apparent m.f.p.'s which are three orders of magnitude smaller than the former. In the highest purity Nb crystals ($RRR \sim 10^4$) we observed ballistic propagation of the phonons over distances ~ 1 cm and quasiparticle diffusion with a m.f.p. of ~ 10 μm . In low purity Nb ($RRR \sim 60$) the m.f.p.'s were 100 μm and 1000 $\text{\AA}$, respectively.

At low temperatures, $T \ll T_c$, the properties of phonons in Nb, in the energy window, $0 < \Omega < 3.1$ meV ($2\Delta_{\text{Nb}}$) should be similar to those in dielectric crystals²⁴. Nb has a higher Debye energy Ω_D than the other superconductors listed in Table 3.1.1. The elevated Debye energy would be expected to lead to long phonon anharmonic lifetimes.²⁵ An enhanced phonon lifetime also increases the effective lifetime of the quasiparticles (see §3.3.2). In §5.5 we suggest that the lifetime of phonons in bulk single crystal Nb appears to be limited by decay mechanisms at the surface of the crystal. We were unable to unambiguously observe thermally limited recombination of quasiparticles in bulk Nb at temperatures below 2 K.

In §3.4 we discuss the response of superconducting tunnel junctions (STJs) to non-equilibrium phonon fluxes from the substrates on which they are fabricated. In the course of this work STJs were used extensively to observe the behaviour of non-equilibrium states generated in the Nb by laser and particle excitations. We also consider the quasi-equilibrium case of a STJ exposed to a black-body distribution of phonons. For the

²²In Sn, In and Pb quasiparticles were shown to arrive in advance of phonon signals over propagation distances of 1–3 mm.

²³Compare Tables 3.2.1 and 5.1.2.

²⁴The condensation of electrons, at the Fermi surface, into bound Cooper Pairs prevents phonons ($\Omega < 2\Delta$) scattering from them.

²⁵The anharmonic decay rate of phonons is proportional to $(\Omega_D)^{-5}$ (see §3.3.8). The Debye temperatures of a number of superconductors are listed in Table 3.3.1.

purposes of the analysis of the experimental work detailed in Chapter 6 it is important to quantify the response of a STJ in both these situations.

3.2. EXPERIMENTALLY DETERMINED PARAMETERS FOR NIOBIUM

Parameter	Value	Comments [Ref.]
v_F Fermi velocity of electrons	$5 \times 10^5 \text{ ms}^{-1}$	Average value used in this work
	$3 \times 10^5 \text{ ms}^{-1}$	[32]
	$5.1 \times 10^5 \text{ ms}^{-1}$	R.m.s. value obtained from $H_{c2}'(1)$ [33]
	$6.2 \times 10^5 \text{ ms}^{-1}$	[34]
	$3 \times 10^5 \text{ ms}^{-1}$	[35]
$N(0)$ Electron density of states of single spin at Fermi surface	$31.7 \times 10^{21} \text{ states eV}^{-1}\text{cm}^{-3}$	See ref. 23 in Kaplan [36]
Θ_D Debye Temperature	275 K	Average value used in this work
	$271 \pm 5 \text{ K}$	120 neV L phonons at 4.2 K [37].
	277 K	Ultrasonic Measurements [38]
ξ_0 Intrinsic Coherence Length	$380 \text{ \AA}$	[39]
λ_L London Penetration Depth	$390 \text{ \AA}$	[39]

Table 3.2.1 Materials data for Nb.

We list the values for a number of experimentally determined parameters for Nb in Table 3.2.1. For calculations in this work we have used the first value listed for each parameter.

3.3. QUASIPARTICLE-PHONON INTERACTION TIMES

3.3.1. Introduction

In this sections we will review the description of the characteristic interaction rates in a superconductor following the work of Kaplan [36]. We have evaluated these rates for Nb. The results are summarised in Fig. 3.3.4 in §3.3.8.

The quasiparticle and phonon systems of a superconductor are coupled. At thermal equilibrium temperature T , the phonon population is described by Bose–Einstein statistics and the quasiparticle population by Fermi–Dirac statistics. The occupancies are given by

$$f(E) = \frac{1}{1 + e^{\frac{(E-\mu)}{kT}}} \text{ (Fermions)}, \quad n(\Omega) = \frac{1}{e^{\frac{\Omega}{kT}} - 1} \text{ (Bosons)}. \quad [3.3.1, 3.3.2]$$

At phonon energies, $\Omega \ll \Omega_D$, the phonon density of states will be related to the phonon energy as

$$F(\Omega) \sim \Omega^2. \quad [3.3.3]$$

The BCS [40] superconducting density of states (relative to that of the normal state) for quasiparticles is given by,

$$\rho(E) = \text{Re} \left[\frac{|E|}{(E^2 - \Delta^2)^{\frac{1}{2}}} \right], \quad [3.3.4]$$

where the quasiparticle energy E is measured relative to the Fermi energy. This function is zero in the range $0 \leq E < \Delta$, the characteristic energy gap associated with the condensation of electrons into bound Cooper Pairs in the superconducting state.

At thermal equilibrium detailed balance requires that the rate of creation of quasiparticles (phonons) at a given energy must equal their rate of annihilation. Analysis of the coupled systems away from thermal equilibrium requires calculations of the interaction rates responsible for the creation and annihilation processes. They are summarised in Fig. 3.3.1 and labelled with the symbols that will be used through out this work when discussing these processes.

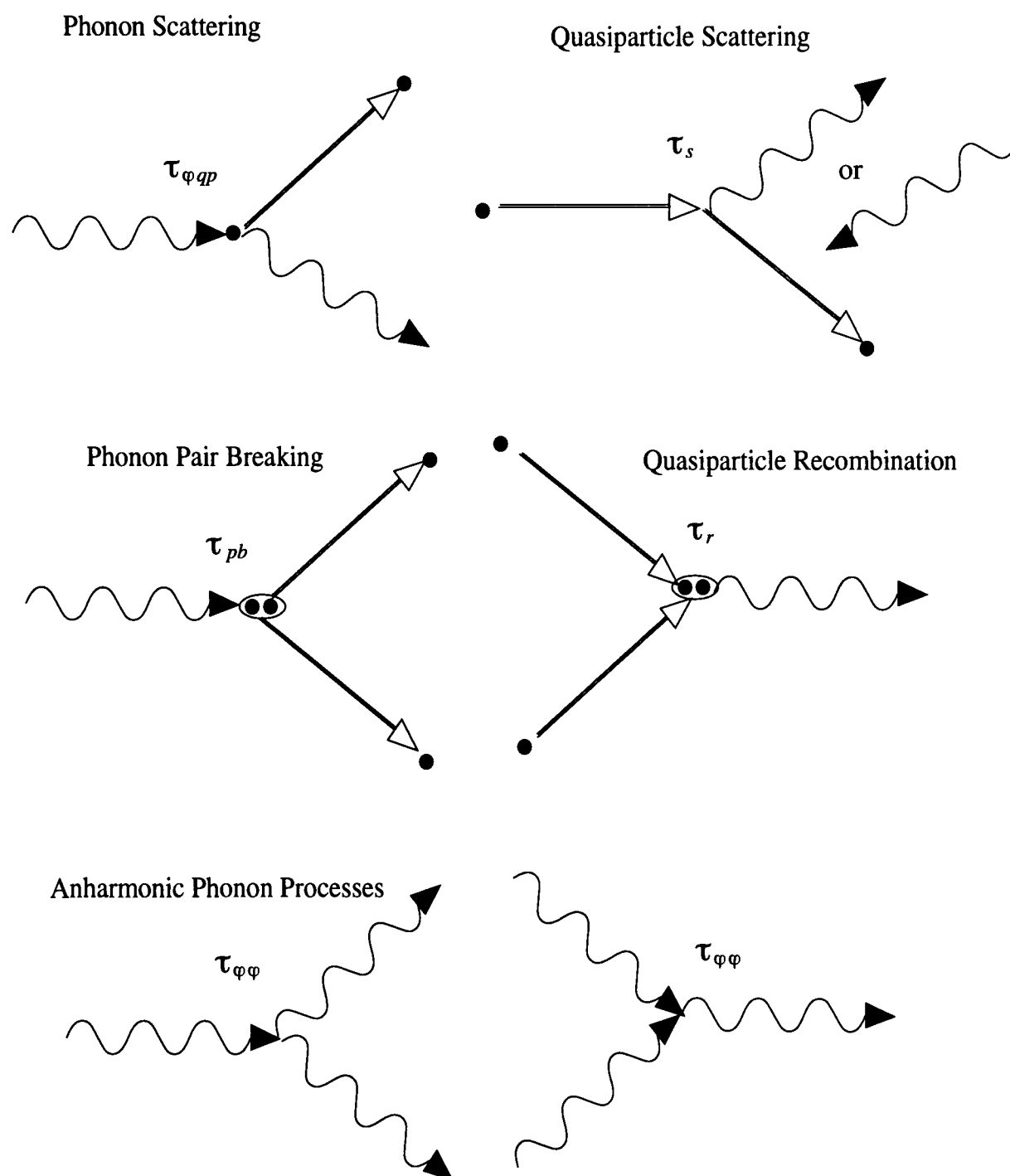


Figure 3.3.1 Schematic of first-order phonon and quasiparticle interactions in a superconductor. Quasiparticle-quasiparticle and impurity scattering are not shown. The processes will be discussed in §3.3.2–3.3.8.

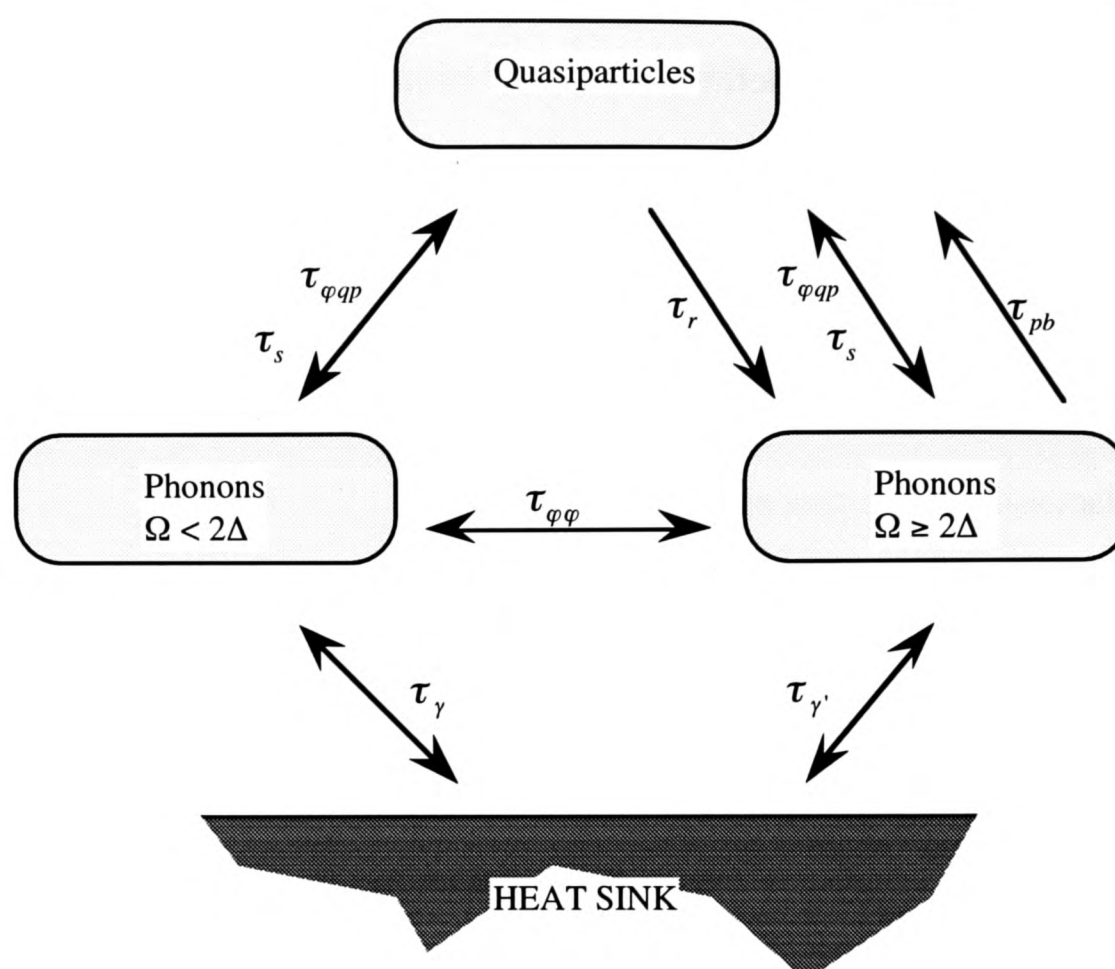


Figure 3.3.2 Schematic of interactions between the electron–phonon system of a superconductor.

Figure 3.3.2 shows schematically how the processes, shown in Fig. 3.3.1, relate to one another in exchanging energy between the phonon and quasiparticle systems. It is appropriate to divide the phonon system into those phonons with energies below and above the superconducting energy gap. Above gap phonons ($\Omega \geq 2\Delta$)²⁶ are able to break Cooper Pairs creating quasiparticles. They are also regenerated by quasiparticle recombination. As a consequence the propagation of non–equilibrium distributions of quasiparticles takes place as a mixed mode of quasiparticles and 2Δ phonons. This is discussed in §3.3.3.

In making the calculations in §3.3.2–3.3.7 we have followed the conventions used by Kaplan *et al.* They used the Eliashberg formulation which gives the imaginary part of the quasiparticle energy (hence the inverse quasiparticle lifetime) in terms of the electron–phonon coupling function $\alpha^2(\Omega)F(\Omega)$. The inverse lifetime can be separated into the inverse lifetimes for recombination and for inelastic quasiparticle scattering [41].

²⁶The energy of a phonon must be twice the superconducting energy gap Δ since two quasiparticles must be created, each with an energy $E \geq \Delta$ when a Cooper pair is broken.

	T_c (K)	Θ_D (K)	Δ (meV)	τ_0^{qp} (ns)	τ_0^{ph} (ps)
Nb	9.2	275	1.515	0.149	4.17
Pb	7.19	105	1.350	0.196	34.0
Sn	3.75	200	0.590	2.30	110
In	3.4	108	0.540	0.799	169
Al	1.19	428	0.180	110 ²⁷	242

Table 3.3.1. Parameters for a number superconductors. Taken from Ref. [36] unless otherwise indicated.

The times of the various quasiparticle–phonon interactions are expressed in terms of the characteristic times τ_0^{qp} , τ_0^{ph} . These are given by

$$\tau_0^{qp} = \frac{Z_1(0)\hbar}{2\pi b(k_B T_c)^3}, \quad \left[\frac{1}{\tau_0^{qp}} \propto T_c^3 \right], \quad [3.3.5]$$

$$\tau_0^{ph} = \frac{\hbar N_{ion}}{4\pi N(0)\alpha_{av}^2 \Delta(0)}, \quad \left[\frac{1}{\tau_0^{ph}} \propto \Delta \propto T_c \right]. \quad [3.3.6]$$

These times correspond to the approximate lifetimes of electrons and phonons in the normal state ($T = T_c$). The quantity $Z_1(0)$, in Eq. 3.3.5, is the electron renormalization factor²⁸. $Z_1(0) = (1+\lambda)$ where λ is the electron-phonon coupling strength. The parameter b is the assumed constant of proportionality in the identity $\alpha^2(\Omega)F(\Omega) \equiv b\Omega^2$ where $\alpha^2(\Omega)$ is the square of the matrix element of the electron-phonon interaction and $F(\Omega)$ is the phonon density of states. The magnitude of $\alpha^2(\Omega)F(\Omega)$ can be estimated, for example, from tunnelling measurements [44]. In Eq. 3.3.6, N_{ion} is the ion number density, $N(0)$ is the single spin electron density of states at the Fermi surface and α_{av}^2 is the average value of α^2 .

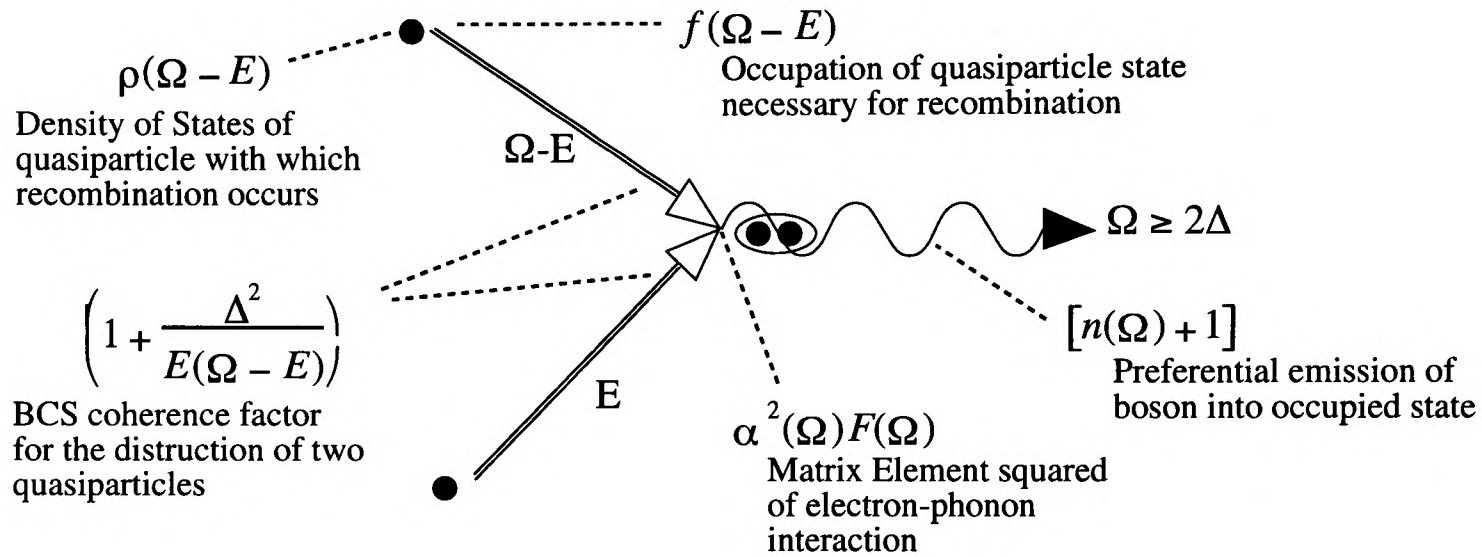
²⁷This value is about 25% of that quoted by Kaplan. The revised estimate is derived from experimental measurements [41–43].

²⁸This factor appears because Kaplan's calculations were made using the Bloch states for the electrons. The fraction of the initial quasiparticle state in this Bloch state is given by the wave–function renormalisation $(Z_1(E))^{-1/2}$. The remaining parts of the state consist of the superposition of virtually scattered electrons and phonons. The energy dependence of this factor is assumed small and consequently ignored.

3.3.2. Quasiparticle Recombination

The rate at which a quasiparticle of energy E recombines with any other is given by

$$\tau_r^{-1}(E) = \frac{2\pi}{\hbar Z_1(0)[1 - f(E)]} \int_{E+\Delta}^{\infty} d\Omega \alpha^2(\Omega) F(\Omega) \rho(\Omega - E) \left(1 + \frac{\Delta^2}{E(\Omega - E)}\right) [n(\Omega) + 1] f(\Omega - E) \quad [3.3.7]$$



Equation 3.3.7 gives the rate of recombination of a quasiparticle of energy E with another of energy $(\Omega - E)$ with the emission of a phonon of energy Ω and the creation of a Cooper pair.

The BCS coherence factor $\left(1 + \frac{\Delta^2}{E(\Omega - E)}\right)$ appears because of the nature of the electron–electron interactions in the superconducting state and the symmetries in the sum of the matrix elements connecting the two quasiparticle states. The sign of the second term is positive for the creation or annihilation of two quasiparticles and negative in the case of a single quasiparticle scattering by a phonon into a new quasiparticle state.

The factor $[1 - f(E)]^{-1}$ represents an enhancement of the decay rate due to the Pauli–principle blocking of the back–scattering of other quasiparticles into the occupied quasiparticle state of energy E .

Making use of the identity

$$[1 - f(E)]^{-1} [n(\Omega) + 1] f(\Omega - E) \equiv [f(\Omega - E) + n(\Omega)], \quad [3.3.8]$$

and rewriting this in terms of τ_0^{qp} and using the approximation

$$\alpha^2(\Omega)F(\Omega) = b\Omega^2, \quad [3.3.9]$$

gives

$$\tau_r^{-1}(E) = (\tau_0^{qp})^{-1} (k_B T_c)^{-3} \int_{E+\Delta}^{\infty} d\Omega \Omega^2 \rho(\Omega - E) \left(1 + \frac{\Delta^2}{E(\Omega - E)} \right) [f(\Omega - E) + n(\Omega)]. \quad [3.3.10]$$

In the low temperature limit $T \ll T_c$ and for energies close to the energy gap, this equation takes the form

$$\tau_r^{-1}(E) = (\tau_0^{qp})^{-1} \pi^{\frac{1}{2}} \left(\frac{2\Delta(0)}{k_B T_c} \right)^{\frac{5}{2}} \left(\frac{T}{T_c} \right)^{\frac{1}{2}} e^{-\left(\frac{\Delta(0)}{k_B T} \right)}. \quad [3.3.11]$$

We can write this in terms of the thermal density of quasiparticles $N(T)$ where ²⁹

$$N(T) = 4N(0) \int_0^{\infty} dE f(E) \rho(E), \quad [3.3.12]$$

$$N(T) = 2N(0) \left(2\pi k_B T \Delta(0) \right)^{\frac{1}{2}} e^{-\left(\frac{\Delta(0)}{k_B T} \right)}, \quad [3.3.13]$$

giving

$$\Gamma_r = \tau_r^{-1} = RN(T), \quad [3.3.14]$$

where R is a constant for a given material. The definition of τ_r adopted here is as used by Gray [45] and Gray, Long and Adkins [46] and as implied by the definition of τ_r^{-1} given in Eq. 3.3.14. The rate is that with which a given quasiparticle recombines with all others. The experimentally observed recombination rate is twice this value since each recombination process removes two quasiparticles.

3.3.3. Effective Quasiparticle Recombination Time

The apparent quasiparticle lifetime in a superconductor will be enhanced over that calculated in the previous section due to the possibility that the recombination phonons break Cooper Pairs in the superconductor regenerating the quasiparticles [47, 48]. The enhancement factor is related to the probability of a 2Δ phonon being lost from the

²⁹The factor of 4 is present in order to include both electron spin states and the hole and electronlike branches.

superconductor before it is able to pair break. In Nb the pair breaking time ($\tau_{pb} \sim \tau_0^{ph}$) is very short (4.17 ps) and corresponds to a phonon m.f.p. of ~ 100 Å. In a bulk superconductor the phonon loss rate will be given by the anharmonic decay time ($\tau_{\phi\phi}$) for a phonon of energy $\Omega = 2\Delta$ ³⁰. In §3.3.9 we calculate this time to be 970 ns. The observed recombination time, τ_{eff} , is given in terms of the other times quoted as

$$\tau_{eff} = \frac{\tau_r}{2} \left(1 + \frac{\tau_{\phi\phi}}{\tau_{pb}} \right), \quad [3.3.15]$$

where the additional factor of 2 emphasises that each recombination actually removes two quasiparticles. In bulk Nb the lifetime of the quasiparticles is enhanced by $\sim 10^5$ by this effect.

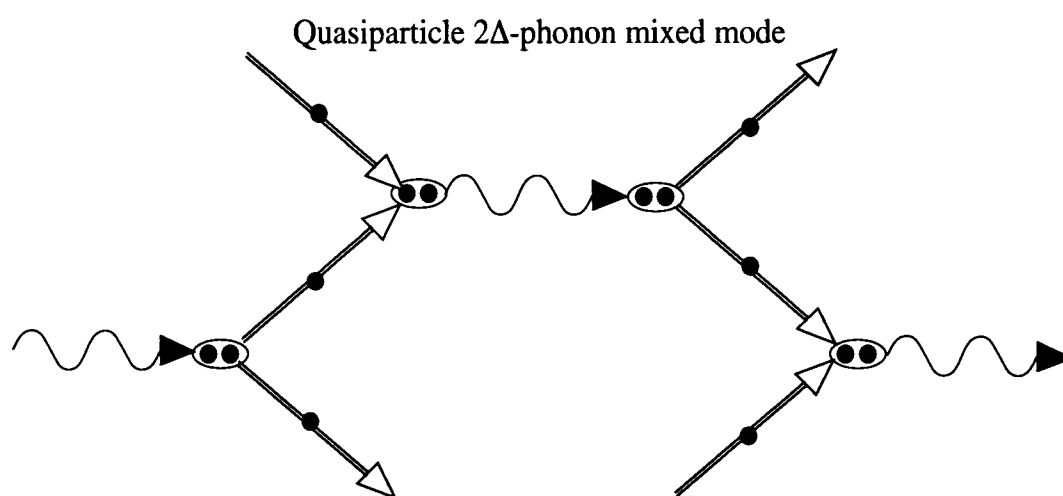


Figure 3.3.3 Schematic representing the propagation of quasiparticles in the mixed-mode.

The propagation of non-equilibrium distributions of quasiparticles will take place as a mixed-mode of quasiparticles and 2Δ -phonons as shown schematically in Fig. 3.3.3 (Note that the same two quasiparticles will not necessarily recombine). The ratio of the energy (of the mixed-mode) in quasiparticles against phonons will be given by

$$\frac{E_{qp}}{E_{\phi=2\Delta}} = \frac{\tau_r}{\tau_{pb}}. \quad [3.3.16]$$

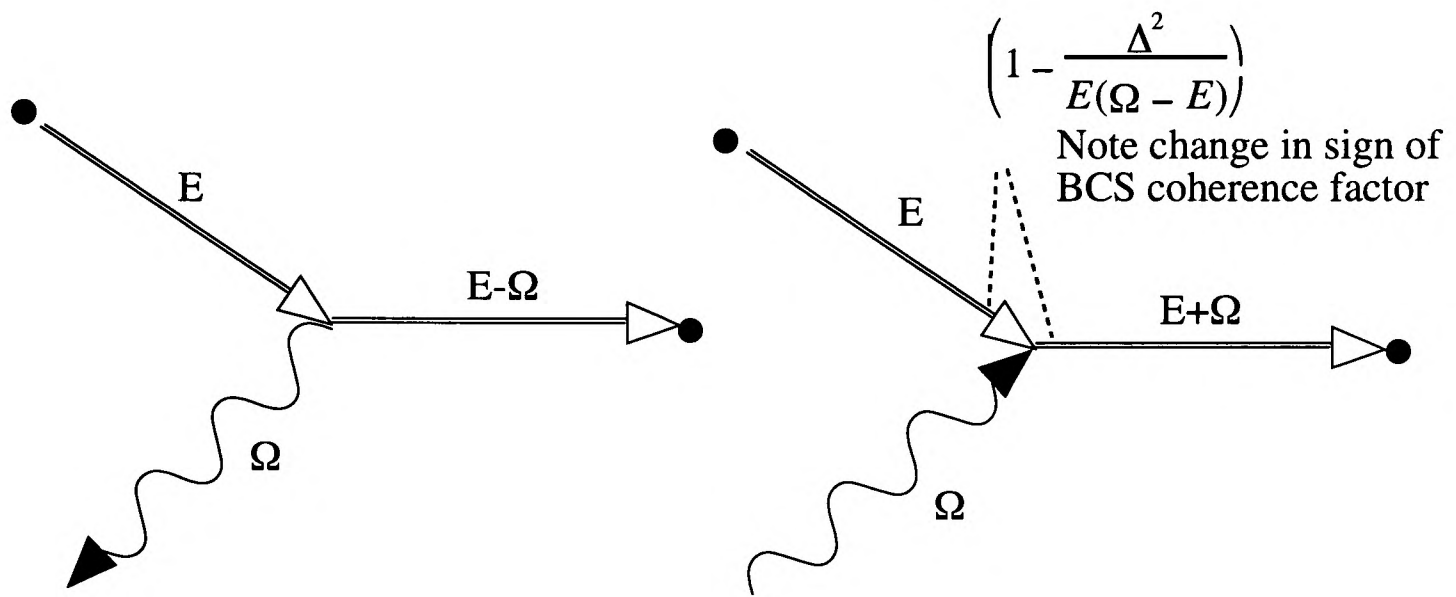
In Nb this ratio only becomes less than 1 for quasiparticle densities corresponding to a temperature of $T \gtrsim 0.9 T_c$ and at temperatures below this the propagation of this mixed-mode will be determined solely by the quasiparticles because of the $e^{-\Delta/k_B T}$ factor.

³⁰In thin films the phonon loss rate will be the sum of the anharmonic decay rate and the rate of loss of the phonons from the film into the substrate.

3.3.4. Quasiparticle Scattering

The inelastic scattering rate τ_s^{-1} for a quasiparticle energy E is the sum of two terms and is given by

$$\tau_s^{-1}(E) = (\tau_0^{qp})^{-1} (k_B T_C)^{-3} \left[\int_0^{E-\Delta} d\Omega \Omega^2 \rho(E-\Omega) \left(1 - \frac{\Delta^2}{E(E-\Omega)} \right) [f(\Omega-E) + n(\Omega)] \right. \\ \left. + \int_0^\infty d\Omega \Omega^2 \rho(\Omega+E) \left(1 - \frac{\Delta^2}{E(E+\Omega)} \right) [f(\Omega+E) + n(\Omega)] \right] \quad [3.3.17]$$



The two terms in Eq. 3.3.17 correspond to the processes of scattering by phonon emission and absorption. The first term (emission) is reduced as the temperature is lowered. The scattering is peaked for final states near the energy gap (due to the form of the quasiparticle density of states), and the increase in the energy gap $\Delta(T)$ leads to a reduction in final state phonon densities ($0 < \Omega \leq E-\Delta$). The second term for phonon absorption in general makes a smaller contribution to the integral and is reduced as the temperature is lowered due to the reduction in the number of phonons available for scattering. This term is important only for quasiparticles at the gap edge ($E = \Delta$) where phonon emission is blocked.

At low temperatures this rate has the form

$$\tau_s^{-1}(E) = (\tau_0^{qp})^{-1} (k_B T_c)^{-3} \left\{ \frac{1}{3} \left[\left(\frac{E}{\Delta} \right)^2 - 1 \right]^{\frac{3}{2}} + \frac{5}{2} \left[\left(\frac{E}{\Delta} \right)^2 - 1 \right]^{\frac{1}{2}} - \frac{\Delta}{2E} \left[1 + 4 \left(\frac{E}{\Delta} \right)^2 \right] \ln \left\{ \frac{E}{\Delta} + \left[\left(\frac{E}{\Delta} \right)^2 - 1 \right]^{\frac{1}{2}} \right\} \right\}. \quad [3.3.18]$$

For quasiparticle energies several times Δ , the scattering rate varies as $(E/\Delta)^3$.

3.3.5. Quasiparticle–Quasiparticle and Impurity Scattering

Lancashire [49] has pointed out the importance of electron-electron scattering processes particularly in Al although Kaplan *et al.* comment that additional corrections in the calculation make an estimate of this effect difficult. Eisenmenger and Dayem [50] and Gray [45] have suggested that for quasiparticle energies above 3Δ additional decay processes become possible where the quasiparticle can relax by directly breaking a Cooper pair. Kaplan states that the relative rates for phonon emission and direct electron-electron scattering at zero temperature are proportional to E_{qp}^3/Ω_D^2 and E_{qp}^2/E_f respectively. This implies that the relative rate τ_s/τ_{e-e} at an energy of $E_{qp} = 3\Delta(0)$ is of order 0.18 for Nb (with phonon emission becoming increasingly more dominant at higher energies due to the additional phase space available).

The rate for the quasiparticle-quasiparticle scattering process has been estimated by Narayanamurti *et al.* [27]. For an electron at the Fermi energy the relaxation rate in the normal state is given by

$$\tau_{e-e}^{-1}(0) = 1.6 \times 10^6 T_c^2 \text{ s}^{-1} \text{K}^{-2}. \quad [3.3.19]$$

In the superconducting state this rate is further reduced as the density of quasiparticles giving

$$\tau_{qp-qp}^{-1} \sim \tau_{e-e}^{-1} \left(\frac{T}{T_c} \right)^{\frac{1}{2}} e^{-\left(\frac{\Delta(0)}{k_B T} \right)}. \quad [3.3.20]$$

For Nb at 1 K this gives a rate of order 1.0 s^{-1} , which is negligible compared to τ_r and τ_s .

The impurity scattering rate may be estimated in a free electron model as

$$\tau_i^{-1}(0) = \frac{ne^2\rho}{m}, \quad [3.3.21]$$

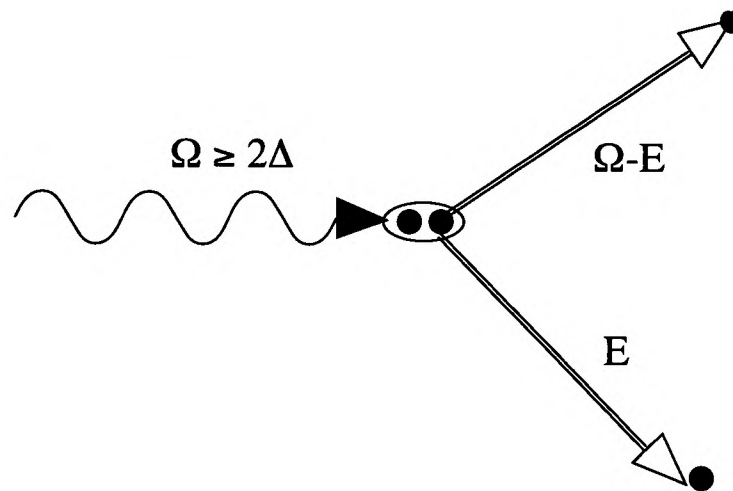
where n is the density of conduction electrons and ρ the resistivity. For Nb, $n = 5.56 \times 10^{28} \text{ m}^{-3}$ and $\rho_{300} = 12.5 \times 10^{-8} \Omega\text{m}$.

Direct measurements were made of the residual resistance ratio of the lower purity Nb crystals, using the eddy-current method, at 10 K giving $\text{RRR} \sim 10$ (This was improved ~ 60 by outgassing). The value for the higher purity samples was taken from an estimate by Schulze who supplied the crystals ($\text{RRR} \sim 10^4$). This is discussed further in §4.2.1. For the higher purity crystals the impurity scattering rate can be estimated as $\tau_i^{-1} = [51 \text{ ps}]^{-1}$. This rate exceeds both τ_r^{-1} and τ_s^{-1} at all temperatures.

3.3.6. Phonon Pair Breaking

This process is the inverse of the quasiparticle recombination process and contains the same coherence factor. The rate is given by

$$\tau_{pb}^{-1}(\Omega) = (\tau_0^{ph})^{-1} \frac{1}{\pi\Delta(0)} \int_{\Delta}^{\Omega-\Delta} dE \rho(E) \rho(\Omega-E) \left(1 + \frac{\Delta^2}{E(\Omega-E)}\right) [1 - f(E) - f(\Omega-E)]. \quad [3.3.22]$$



For $T < T_c/2$ and energies near 2Δ this process tends to the value

$$\tau_{pb}(2\Delta) = \tau_0^{ph} \quad [3.3.23]$$

Pair breaking is evidently discontinuous as the phonon energy is lowered and will only occur for $\Omega \geq 2\Delta$. This fact leads to the discontinuity in the phonon absorption coefficient derived by Bobetic [51]. For phonons propagating in a superconductor we expect the nature of the transport to be different for the two groups of phonons $\Omega < 2\Delta$ and $\Omega \geq 2\Delta$.

For the first group there will be little interaction with the quasiparticles at low temperatures and these phonons will propagate ballistically, limited only by impurity scattering, with velocities equal to the sound velocity.

3.3.7. Phonon–Quasiparticle Scattering

This is the inverse process of the quasiparticle scattering of §3.3.3. The coherence factors are the same and the rate is given by

$$\tau_{\phi qp}^{-1}(\Omega) = (\tau_0^{ph})^{-1} \frac{2}{\pi\Delta(0)} \int_{\Delta}^{\infty} dE \rho(E) \rho(\Omega + E) \left(1 - \frac{\Delta^2}{E(\Omega + E)}\right) [f(E) - f(\Omega + E)], \quad [3.3.24]$$

The temperature dependence of this process is determined by the reduction in quasiparticle numbers as the temperature is lowered. For $\Omega \ll k_B T$ and $\Omega \ll \Delta(T)$ the expression gives

$$\tau_{\phi qp}^{-1}(\Omega) = (\tau_0^{ph})^{-1} \frac{2\Omega\pi}{\Delta(0)} f(\Delta(T)) \quad [3.3.25]$$

3.3.8. Summary of Temperature Dependence of Characteristic Times for Phonons and Quasiparticles

Figure 3.3.4 shows the temperature dependence of the lifetimes of the quasiparticles and phonons of given energies in Nb where the characteristic times τ_0^{qp} and τ_0^{ph} are those from Kaplan (see Table 3.3.1). The energies are chosen to illustrate point brought out in the text.

The recombination time and scattering time for a quasiparticle of energy $\Delta(T)$ are strongly temperature dependent. The loss rate of a quasiparticle, of a given energy, due to recombination decreases as the temperature is lowered due to the reduction in the number of other quasiparticles with which to recombine. At the gap energy the scattering rate of a quasiparticle to another energy is determined only by the probability of absorption of a phonon since it is prevented from emitting a phonon.

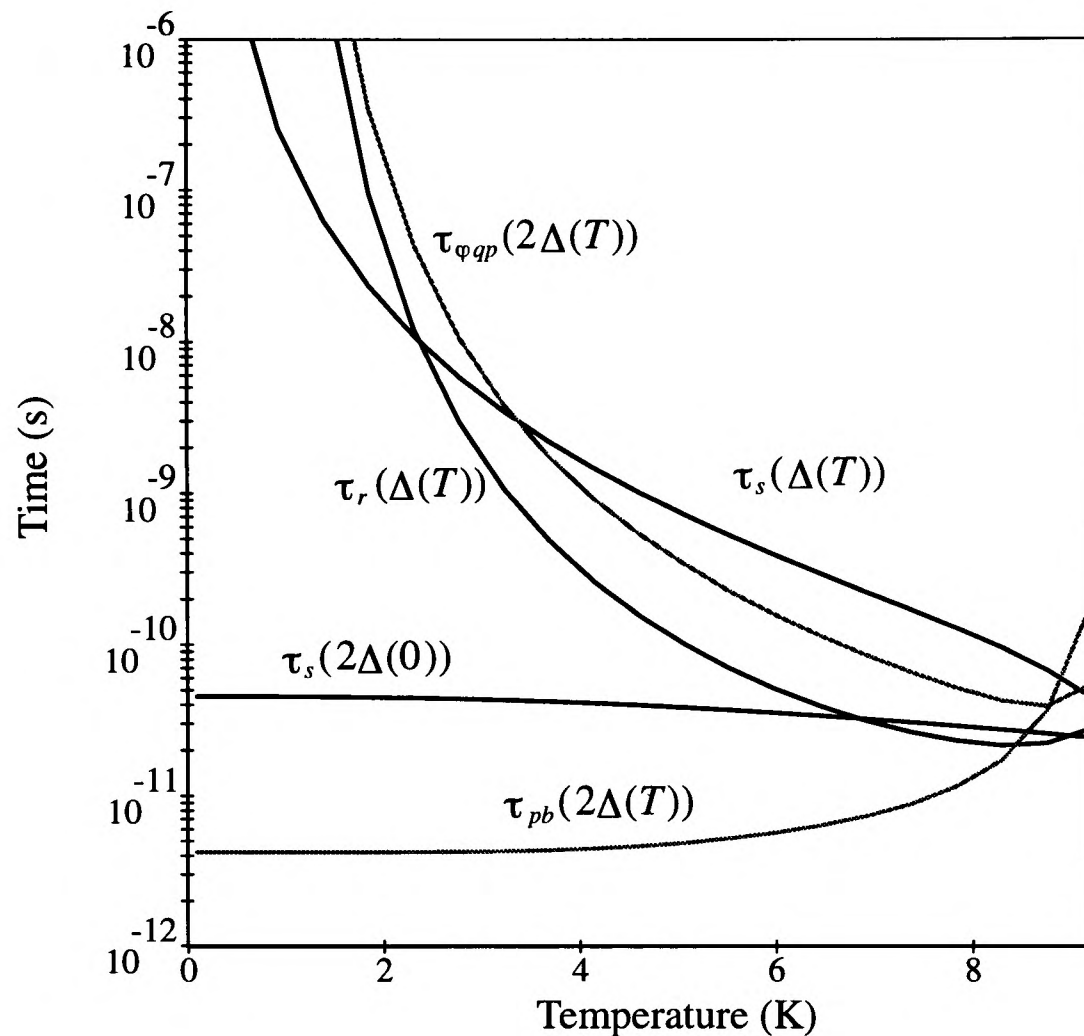


Figure 3.3.4 Lifetimes of quasiparticles (black lines), and phonons (grey lines) of given energy in Nb as a function of temperature.

The scattering rate of quasiparticles at 2Δ is nearly independent of temperature since the first integral in Eq. 3.3.17 now dominates, that corresponding to phonon emission. The residual temperature dependence arises from the variation in the gap energy $\Delta(T)$.

At low temperatures a phonon with $\Omega \geq 2\Delta$ is much more likely to break a Cooper pair than scatter from a quasiparticle.

3.3.9. Phonon Decay Rates

In addition to processes involving quasiparticle interactions, other mechanisms can contribute to the loss of phonons from a superconductor. The anharmonic phonon decay rate has been studied by Orbach et al. [52] and by Klemens [53] for high energy phonons $\Omega \gg k_B T$. In the experimental work presented here the phonon energies of interest are of order $2\Delta_{Al} - 2\Delta_{Nb} \simeq 5\text{-}35\text{ K}$ which satisfies this condition.

The process of interest is the decay $\Omega_1 \rightarrow \Omega_2 + \Omega_3$ where the conservation rules for energy and momentum imply $\phi_L \leftrightarrow \phi_T + \phi_T$ or $\phi_L \leftrightarrow \phi_L + \phi_T$. The net decay rate used by Klemens and given by Langenberg [54] is

$$\tau_{\phi\phi}^{-1}(\Omega) = \frac{3\pi\hbar}{2^{\frac{5}{2}}} \gamma^2 \frac{c_L}{c_T} \frac{\Omega^5}{Mc_L^2\Omega_D^3}.$$

[3.3.26]

Here $c_{L,T}$ are the sound velocities for longitudinal and transverse phonons, M is the atomic mass, Ω_D is the Debye energy, and γ is the Gruneissen constant ($\simeq 2$ for metals). Equation 3.3.26 gives $\tau_{\phi\phi}$ of 64 ns at $\Omega = 2\Delta_{Nb}$ (The decay times at other energies go as Ω^{-5}).

A more detailed theoretical analysis of the spontaneous decay of LA phonons, in terms of the second and third-order elastic constants, was performed by Tamura [55]. Tamura’s calculations showed that the estimates of the LA lifetime in Si and Ge of Klemens were an order of magnitude too short. Using the room temperature third-order elastic constants for Nb determined by Chang [56] (see Table 3.3.2) we estimate the decay times of phonons ($\Omega = 2\Delta_{Nb}$) via the processes $\phi_L \leftrightarrow \phi_T + \phi_T$ or $\phi_L \leftrightarrow \phi_L + \phi_T$ are 970 ns and 107 μ s respectively.

c111	c112	c123	c144	c155	c456	c_L	c_T	ρ
10^{10} dyne cm ⁻²						(km s ⁻¹)	(km s ⁻¹)	(gcm ⁻³)
-2560	-1140	-467	-343	-168	+137	5.118	2.135	8.57

Table 3.3.2 Third order elastic constants in single crystal Niobium from Chang [56, 57]. Also shown are the phonon velocities, and density for Nb used in the calculation of the phonon decay rates. Second-order elastic constants used in calculation are from Weber [37] and are quoted in Table 6.2.1.

It is apparent that the decay of LA phonons is dominated by that to two TA phonons and that Tamura’s calculation gives a lifetime over an order of magnitude longer than that of Klemens.

	Energy (μeV)	Decay Time (Klemens Model)	Decay Time (Tamura Model)
$2\Delta_{\text{Al film}}$	400	1.8 ms	24.2 ms
$2\Delta_{\text{Sn film}}$	1150	6.6 μs	123 μs
Δ_{Nb}	1515	2.1 μs	31.1 μs
$2\Delta_{\text{Pb film}}$	2730	110 ns	1.63 μs
$2\Delta_{\text{Nb}}$	3030	64 ns	970 ns

Table 3.3.3 Decay times of L phonons at various energies of interest calculated using the models of Klemens and Tamura. We will use the times from Tamura in this paper unless otherwise specified.

The Klemens and Tamura calculations were made assuming zero temperature. The effect of a thermal distribution of phonons at temperature T would be to enhance the decay rate by a factor

$$(1 + n(\Omega_2))(1 + n(\Omega_3)) + U(\Omega, T), \quad [3.3.27]$$

where Ω_2 and Ω_3 are the energies of the decay product phonons (which will most probably have energies $\Omega_2 = \Omega_3 = \Omega/2$). $U(\Omega, T)$ represents the contribution from $\phi + \phi \leftrightarrow \phi(\Omega)$ processes. For $\Omega \sim 2\Delta_{\text{Nb}}$ Eq. 3.3.27 will be ~ 1 at all temperatures of interest. At $\Omega = 2\Delta_{\text{Al}}$ at a temperature of 1 K the first terms of Eq. 3.3.27 have a value ~ 1.3 .³¹

3.4. TUNNEL JUNCTION DETECTOR RESPONSE AND PARAMETERISATION OF DETECTOR SIGNALS

3.4.1. Introduction

In this section we will briefly discuss the behaviour of the tunneling current in a superconducting tunnel junction (STJ) and its variation with bias voltage and temperature.³² We will then go on to look at the response of a STJ to a phonon flux from the substrate on which it is fabricated. We will determine a relation between the size of

³¹A study of the detailed behaviour of non-equilibrium phonons injected into thermal backgrounds has been pursued further by Gaitskell *et al.* [58]

³²This is discussed extensively in the existing literature. See for example Giaever and Megerle [59], Van Duzer [39], Eisenmenger [60] and Gray [61].

the voltage signal, δV , from a current biased STJ, and the phonon flux, $\delta\Phi$, entering it. The sensitivity ($\delta V/\delta\Phi$) is enhanced as the temperature decreases. We also calculate the voltage signal from an STJ (initially at temperature T) when exposed to a quasi-thermal distribution of phonons (represented by a temperature $T+\delta T$). We show, in this case that the apparent sensitivity ($\delta V/\delta T$) of the STJ falls as the temperature decreases.

3.4.2. Superconducting Tunnel Junction Characteristics

The current flowing in a STJ, at a bias voltage, V , is equal to

$$I(V, T) = \frac{1}{R_{nn}e} \int_{-\infty}^{\infty} dE \rho_1(E) \rho_2(E+V) [f(E) - f(E+V)]. \quad [3.4.1]$$

For a homogeneous junction (where $\Delta = \Delta_1 = \Delta_2$), at voltages less than 2Δ and at temperatures, $k_B T \ll eV$, so that $f(E+eV) \ll 1$, an approximate expression for the thermal quasiparticle current is

$$I(V, T) = \frac{1}{R_{nn}e} \rho(E+V) 2 \int_{\Delta}^{\infty} dE \rho(E) f(E), \quad \rho(E) = \frac{E}{[E^2 - \Delta^2]^{\frac{1}{2}}}. \quad [3.4.2]$$

The first density of states term, $\rho(E+V)$, has been brought outside the integration since it is a slowly varying function in the integration range. It can be approximated ~ 1 . The integral term in Eq. 3.4.2 is simply the thermal density of excitations $N(T)$ divided by $4N(0)$ given in Eq. 3.3.12. We can define the tunneling rate for the quasiparticles such that

$$I(T) = 2e\Gamma_{tun} N(T) vol, \quad [3.4.3]$$

where vol is the tunneling volume and the factor of two takes account of tunneling of the hole and electron-like excitations. Hence the tunneling rate is ³³

$$\Gamma_{tun} = \left[\epsilon e^2 N(0) R_{nn} (vol) \right]^{-1}. \quad [3.4.4]$$

³³If the density of electron states (single spin) at the Fermi surface, $N(0)$, is expressed in [states $eV^{-1} vol^{-1}$], the factor e should appear only once.

In the case we have been considering ϵ is 4. However, for tunneling between non-identical superconductors it can become a function of both the quasiparticle energy and the bias voltage [62]. We will not consider this case further.

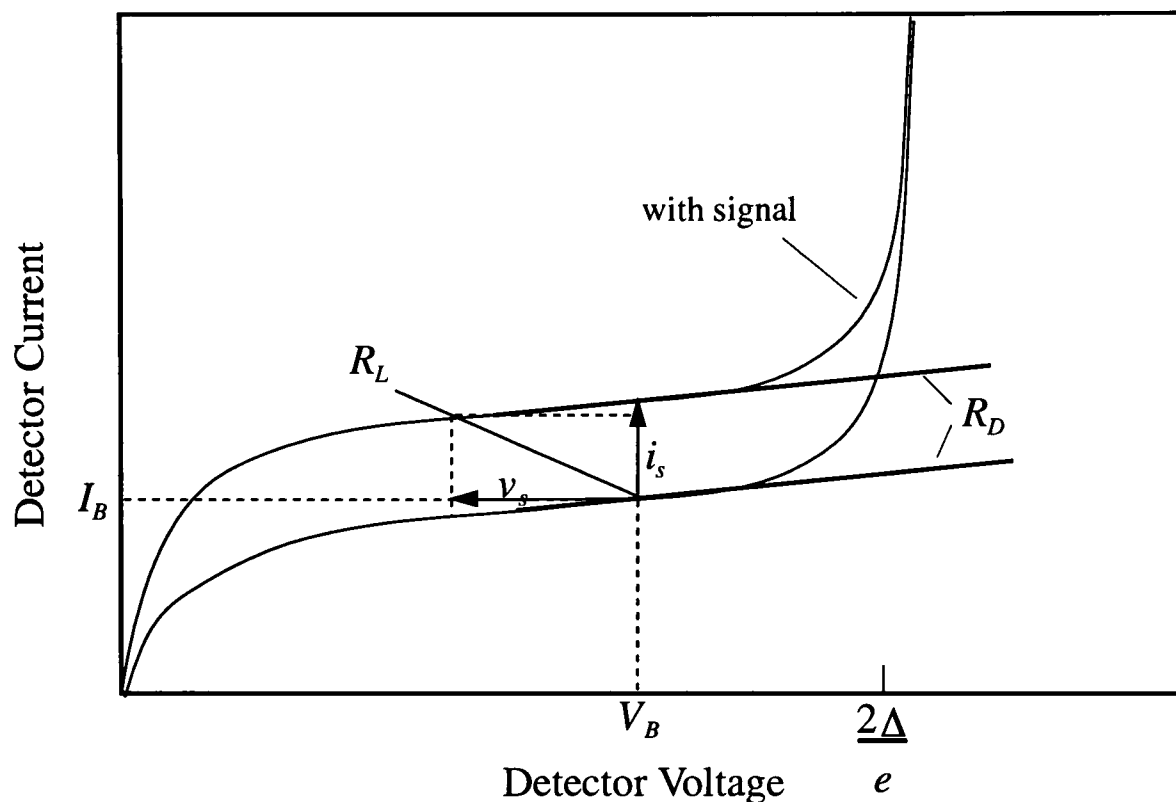


Figure 3.4.1 Load line consideration for STJ detector signal in a current bias circuit. Symbol definitions are in the text.

The electrical response of a constant current biased (I_B) STJ to a pulsed injection of quasiparticles is summarised in Fig. 3.4.1. The increase in the quasiparticle density within the STJ films effectively shifts the IV characteristic to one corresponding to a slightly higher temperature. The increase in the thermal tunneling is described by the short-circuit signal current, i_s . The effect of the constant current bias circuitry is, however, to make this current appear as a shift in the apparent voltage bias (V_B) by an amount, v_s . The signal voltage is then given by the relation

$$v_s = -\left(\frac{1}{R_D} + \frac{1}{R_L}\right)^{-1} i_s. \quad [3.4.5]$$

In practice the load resistor, R_L , of the biasing circuitry is chosen to be very much greater than the dynamic impedance, R_D , of the STJ and so this relationship simplifies to

$$v_s = -R_D i_s. \quad [3.4.6]$$

At large injections of quasiparticles, $\delta N_{qp} \sim N_{qp}$ (the thermally generated number) the non-linearities of the I–V characteristic forbid this relationship. However, for the data reported in this work careful attention was paid so as not to enter this regime.

The electronic detector time constant is given by

$$\tau_{RC} = C \left(\frac{R_D R_L}{R_D + R_L} \right), \quad [3.4.7]$$

where C is the junction capacitance (in parallel with the amplifier input and lead capacitances). Typical values, for the STJs, in this work were, a dynamic resistance of 30Ω and a capacitance 3 nF ³⁴ giving a time constant less than 100 ns .³⁵

3.4.3. Relationship Between Voltage Signal from STJ and Non-Equilibrium Phonon Flux Entering Films

We will describe a method for converting the voltage changes observed in a constant current biased STJ, due to a laser or particle excitation in the substrate, into a form more closely related to the processes taking place in the substrate. If non-equilibrium phonons, from an excitation in the substrate, pass from the substrate into the thin films (energy gap, Δ) of the STJ, those phonons with energies $\Omega \geq 2\Delta$ will break Cooper Pairs creating additional quasiparticles. The quasiparticles will tunnel through the junction barrier leading to a change in the voltage bias of the STJ as described in the previous section. The excess quasiparticles will ultimately be lost, due to recombination with the thermal quasiparticle population³⁶. We will show how the instantaneous voltage signal, $\delta V (= v_s$, in previous section) can be converted into a phonon flux rate, $\delta \Phi$.

The quasiparticle number in the films can also be related to an effective temperature rise of the film. This latter concept is most useful if the temperature rise of the film is

³⁴Oxide barriers on Al have a typical capacitance of $6 \mu\text{F cm}^{-2}$. The total of the lead and amplifier input capacitances were $\sim 200 \text{ pF}$.

³⁵ For the Sn–Ox–Sn STJs the high normal state resistances lead to problems with RC effects. Shorting resistors, R_{ll} , were used across the device to reduce the effective electronic time constant. In this case, R_{ll} should be substituted for R_D in Eq. 3.4.7 above. Signal to noise is sacrificed to improve the timing response of the detector.

³⁶Quasiparticle loss will also take place due to their out diffusion down the STJ leads. In addition, their effective lifetime will be enhanced by pair breaking of the recombination phonons (see §3.3.2). We will assume all these effects are taken into account in the quoted lifetime, τ_r .

assumed to be the result of an overall rise in temperature of the substrate with which the junction is in thermal equilibrium. This approach should not be confused with the non-equilibrium T^* and μ^* analyses of Chang and Scalapino [63, 64]. Our analysis can be viewed as simply using the STJ as a thermometer although it still primarily couples to the phonon distribution through the above gap phonons. This will be discussed in more detail in §3.4.4 and §3.4.5.

We will now examine the relationship between the voltage signals, observed in the current biased STJ, and the (non-equilibrium) phonon flux rate entering the detector. If the phonon flux creating excess quasiparticles in the superconducting films of the junction is $\Phi(t)$ then the time development of signal voltage is given by

$$\delta V(t, T) = e\Gamma_{tun} R_D(T) \int_{-\infty}^t dt' \alpha(T, t - t') 2\beta(T, \Omega) \Phi(t') \quad [3.4.8]$$

where T is the equilibrium temperature of the crystal and STJ, $R_D(T)$ is the dynamic resistance of the junction at the bias point, Γ_{tun} is the tunnelling rate of the quasiparticles across the junction, β is the efficiency with which phonons (energy Ω) break Cooper Pairs creating two quasiparticles and $\alpha(T, t)$ is the response function of the STJ.

The coefficient β contains a number of different effects. Clearly, for $\Omega < 2\Delta$, it will be zero due to the cut off in the Pair breaking energy. Above this threshold the number of quasiparticles created will show a non-linear dependence with Ω . This is illustrated in Fig. 3.4.2. In the range $2\Delta < \Omega < 4\Delta$ the phonon will still only create two quasiparticles. By considering the behaviour of the integral in Eq. 3.3.22 it is apparent that the most probable outcome, when a Cooper pair is broken, is for one quasiparticle to have an energy Δ and the other an energy $\Omega - \Delta$. The second quasiparticle will relax rapidly emitting a phonon, energy $\Omega - 2\Delta$ (see §3.3.4). This phonon is unable to break an additional Cooper pair. If $\Omega \geq 4\Delta$ additional pair breaking will take place but the net effect of the mechanism described will be for the STJ to show a 50% efficiency. In the absence of more detailed information on the spectra of phonons we were detecting, we

have taken note of this effect, but assumed that we were in the regime where m (defined in Fig. 3.4.2) ~ 1 .

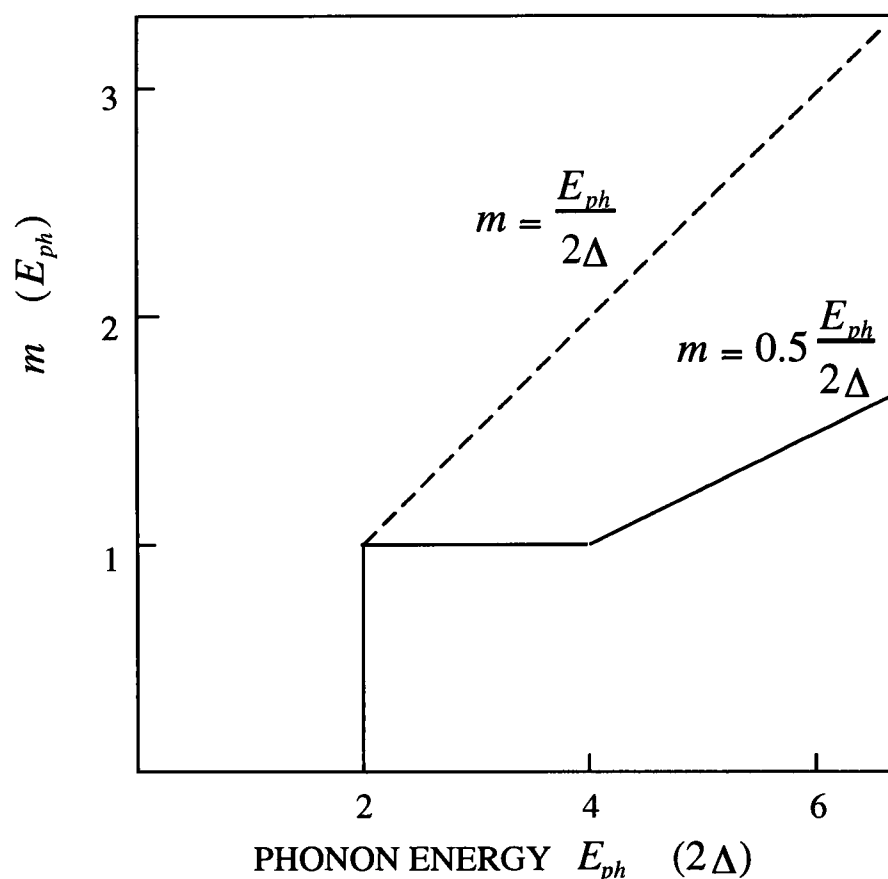


Figure 3.4.2 Detector phonon counting rate m as a function of phonon energy E_{ph} . The parameter $m = 1$ corresponds to the excitation of two quasiparticles by Cooper Pair breaking in the energy range $2\Delta < E_{ph} < 4\Delta$. For higher energies the counting rate is expected to increase as $m = 0.5 E_{ph}/2\Delta$ by additional 2Δ -phonons produced by quasiparticle relaxation in the detector. The dashed line corresponds to a counting rate increasing in proportion to energy with $m = 1$ at $E_{ph} = 2\Delta$. Figure is from Eisenmenger [60] after Forkel.

β also contains a factor related to the probability of an above gap phonon pair breaking before it either decays in the film or leaves the film back into the substrate. The pair breaking rate itself also has a small temperature dependence (see §3.3.6), however for $T < 0.5T_c$ (of the film), this can be assumed to be constant. In the experiments we conducted the film thicknesses were typically in excess of the m.f.p. for pair breaking for the materials used. Again we assume the efficiency is unity.

The transmission probability of phonons from the substrate in to the film will also be dependent on the phonon energies and modes. An adjustment could be made to β using the acoustic mismatch model [54, 65], however, the results will not be significantly altered.

For the purposes of evaluating Eq. 3.4.8 we will assume β is independent of temperature and ~1 bearing in mind that the STJ becomes a phonon counter rather than measuring the total energy in the phonon flux. In the following calculations we will simply use a factor of 2Δ to convert the phonon number back into an energy flux.

$\alpha(T, t-t')$ is a function describing the response of the detector arising from the combined effects of the effective recombination time of the quasiparticles in the STJ films (τ_r) and the electronic RC time constant (τ_{RC}) discussed in §3.4.2. It has the form (see Appendix C)

$$\alpha(t) = \frac{\tau_r}{(\tau_{RC} - \tau_r)} \left[e^{-\frac{t}{\tau_{RC}}} - e^{-\frac{t}{\tau_r}} \right] \quad [3.4.9]$$

Both τ_r and τ_{RC} are function of temperature. In the case where $\Phi(t)$ is a step function, $\Phi(t) = \Phi_0$ ($t \geq 0$), a constant flux rate of phonons, Eq. 3.4.8 becomes:

$$\delta V_0 = e\Gamma_{tun} R_D(T) \tau_r(T) 2\beta \Phi_0, \quad t \gg \tau_{RC}, \tau_r \quad [3.4.10]$$

At a given temperature $R_D(T)$ and $\tau_{Al}(T)$ can be measured directly and Γ_{tun} determined from the normal state resistance of the STJ. Voltage signals can be expressed as phonon energy (or number) fluxes (assuming $\beta \approx 1$).

The total phonon number in a signal can be obtained by the time integration of the voltage signal (in effect the total charge collected) which is then converted using Eq 3.4.8.

$$\int (\beta \Phi) dt = (2e\Gamma_{tun} R_D(T) \tau_r(T))^{-1} \int V(t) dt. \quad [3.4.11]$$

In the cases where we wished to extract variations in the phonon flux which are on time-scales comparable to τ_r , it is necessary to return to the original form of Eq. 3.4.8. Within numerical factors this is a convolution of the functions $\alpha(t)$ and $\Phi(t)$. Using Fourier Transforms (FT) it is possible to show that the phonon flux is given by

$$\Phi(t) = FT^{-1} \left[\frac{FT[\delta V(t')]}{FT[\alpha(t')]} \right]. \quad [3.4.12]$$

Figure 3.4.3 shows the detector response function used to obtain the transformation of the STJ voltage signals pulse in Fig. 5.2.1 to the energy flux pulse in Fig. 5.2.2.

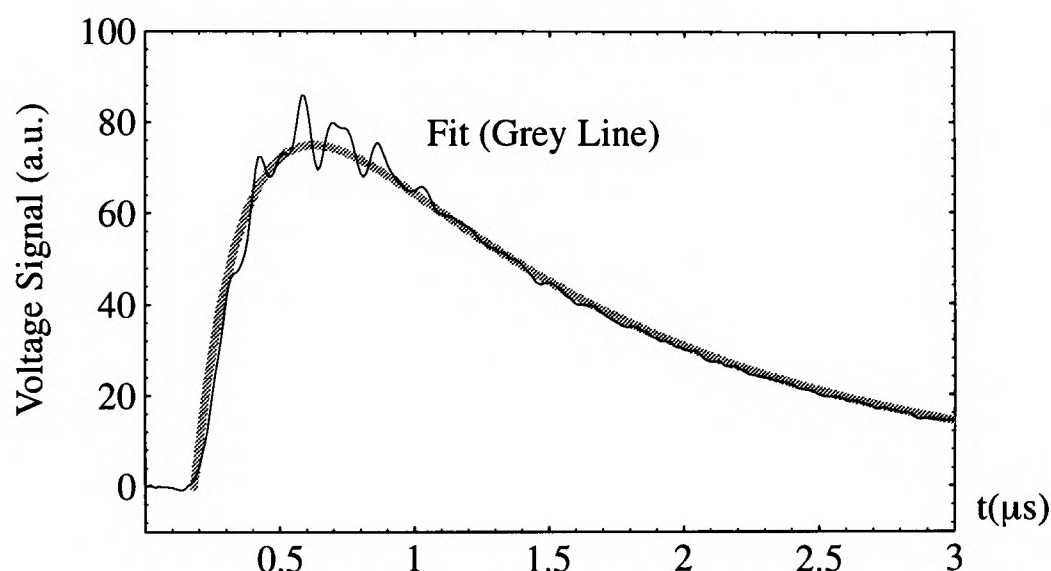


Figure 3.4.3 The detector response function for the STJ used in the ballistic phonon experiment of Fig. 5.2.1. The pulse was obtained by direct illumination of the STJ by the laser. The line fit is $\alpha(t)$ with $\tau_r = 1.30 \mu s$, $\tau_{RC} = 0.20 \mu s$ (and an offset of $0.18 \mu s$).³⁷

3.4.4. Agreement of the Thermal and Dynamic Resistance Models of STJ Response

For a given quasiparticle excess in an STJ the voltage signal (for constant current bias) can be calculated in two ways: (1) considering an effective non-equilibrium temperature rise in the thin film quasiparticle system due to an increase in the number of quasiparticles or (2) considering the effect of the additional tunnelling current in the STJ due to the excess number of quasiparticles. We intend to show these two interpretations are consistent in most regions of junction bias and consequently using a calculation based on the dynamic resistance of the junction is valid.

We can compare the two methods outlined above using the following assumptions. The form of the IV curve, $eV < 2\Delta$ and $kT \ll \Delta$ is

$$I(x, y) = \frac{2\Delta}{R_{nn}e} f(x, y), \quad [3.4.13]$$

$$y = \frac{\Delta}{k_B T}, \quad x = \frac{eV}{2\Delta}, \quad [3.4.14]$$

³⁷File: "Ballistic Cross Xtal VIIIp47"

where $f(x,y)$ can be assumed to take the form given by Van Duzer [39] p87 for a homogenous STJ (This equation is a more complete form of Eq. 3.4.2)

$$f(x,y) = e^{-y}(1+x)^{-\frac{1}{2}}(1+2x)\sinh(xy)K_0(xy), \quad [3.4.15]$$

where K_0 is the zeroth-order modified Bessel function.³⁸

The quasiparticle excess can be related to the temperature by the low temperature approximation³⁹

$$n_{qp} = \alpha y^{-\frac{1}{2}} e^{-y}, \quad \alpha = 2N(0)(2\pi)^{\frac{1}{2}}\Delta, \quad y \gg 1, \quad [3.4.16]$$

and its temperature dependence is given by

$$\frac{\partial n_{qp}}{\partial y} = -n_{qp} \left(1 + \frac{1}{2y} \right) \cong -n_{qp}, \quad y \gg 1. \quad [3.4.17]$$

In addition the tunnelling rate of the quasiparticles is related to the normal state resistance by

$$\Gamma_{tun} = \left[4eN(0)R_{nn}(vol_{film}) \right]^{-1}, \quad vol_{film} = \frac{1}{2} vol, \quad [3.4.18]$$

where vol is the total junction volume and $N(0)$ is density of electron states at the Fermi surface for one spin state in states $\text{eV}^{-1} \text{vol}^{-1}$. The dynamic resistance is given by

$$\frac{1}{R_D} = \frac{\partial I}{\partial V} \Big|_y = \frac{e}{2\Delta} \frac{\partial I}{\partial x} \Big|_y = \frac{1}{R_{nn}} \frac{\partial f}{\partial x} \Big|_y. \quad [3.4.19]$$

We now look at two independent ways of parameterising the behaviour of the STJ IV characteristic when an excess number of quasiparticles, δN , is present over the thermal equilibrium number, N .

³⁸The Bessel functions (of first and third kind) are given by

$$K_0(x) = \frac{\pi}{2} i H_0^{(1)}(ix) \text{ [which will be real for real } x]$$

$$H_0^{(1)}(x) = J_0(x) + iN_0(x) \text{ [latter term is } 0/0 \text{ but is well defined in the limit]}$$

³⁹ In this notation $\tau_r^{-1} = \left(\tau_0^{qp} \right)^{-1} \left(\frac{\pi}{2} \right)^{\frac{1}{2}} \left(\frac{2\Delta}{k_B T_C} \right)^3 y^{-\frac{1}{2}} e^{-y}$

The voltage signal arising from the change in temperature of thin films due to an excess of quasiparticles, δN at a constant current bias is

$$\delta V^{(T)} = \left. \frac{\partial V}{\partial n_{qp}} \right|_I \frac{\delta N}{vol} \cong - \left. \frac{\partial V}{\partial y} \right|_I \frac{\delta N}{n_{qp} vol}. \quad [3.4.20]$$

The partial differential of the voltage with inverse temperature at constant current can be rewritten

$$\left. \frac{\partial V}{\partial y} \right|_I = \frac{2\Delta}{e} \left. \frac{\partial x}{\partial y} \right|_I = - \frac{2\Delta}{e} \left\{ \left. \frac{\partial f}{\partial y} \right|_x \right\} \left\{ \left. \frac{\partial f}{\partial x} \right|_y \right\}^{-1}. \quad [3.4.21]$$

Alternatively if the voltage signal is assumed to arise from the excess current over the dynamic resistance of the junction

$$\delta V^{(R)} = e \Gamma_{tun} R_D \delta N, \quad [3.4.22]$$

$$\delta V^{(R)} = \frac{1}{2e} \left(\frac{\delta N}{N(0)vol} \right) \left(\frac{R_D}{R_{nn}} \right). \quad [3.4.23]$$

The ratio of the two signals is

$$\frac{\delta V^{(R)}}{\delta V^{(T)}} = - \frac{n_{qp}}{4N(0)\Delta} \left\{ \left. \frac{\partial f}{\partial y} \right|_x \right\}^{-1} \quad [3.4.24]$$

Most of the temperature dependence can be isolated in the partial differential on the RHS of Eq. 3.4.24 by the substitution

$$\left. \frac{\partial f}{\partial y} \right|_x = y^{-\frac{1}{2}} e^{-y} \beta(x, \{y\}) = \frac{n_{qp}}{\alpha} \beta(x, \{y\}) \text{ where } \beta \text{ is a weak function of } y \quad [3.4.25]$$

which allows the removal of n_{qp} from the ratio

$$\frac{\delta V^{(R)}}{\delta V^{(T)}} = - \left(\frac{\pi}{2} \right)^{\frac{1}{2}} \frac{1}{\beta(x, \{y\})} \text{ where } \beta(x, \{y\}) = y^{\frac{1}{2}} e^y \left. \frac{\partial f}{\partial y} \right|_x. \quad [3.4.26]$$

For an homogenous STJ with BCS gap $2\Delta = 3.53 k_B T$ the ratio has the form shown in Fig. 3.4.4 for various value of T/T_c . The signal sizes are nearly identical for biases $eV \geq 0.1\Delta$. At low biases the shape of the IV curve means, following an injection of quasiparticles,

assuming that the STJ is biased at a point on a constant IV curve but at a voltage which gives a thermal current equal to the original thermal current minus the excess tunnelling current, is no longer valid.

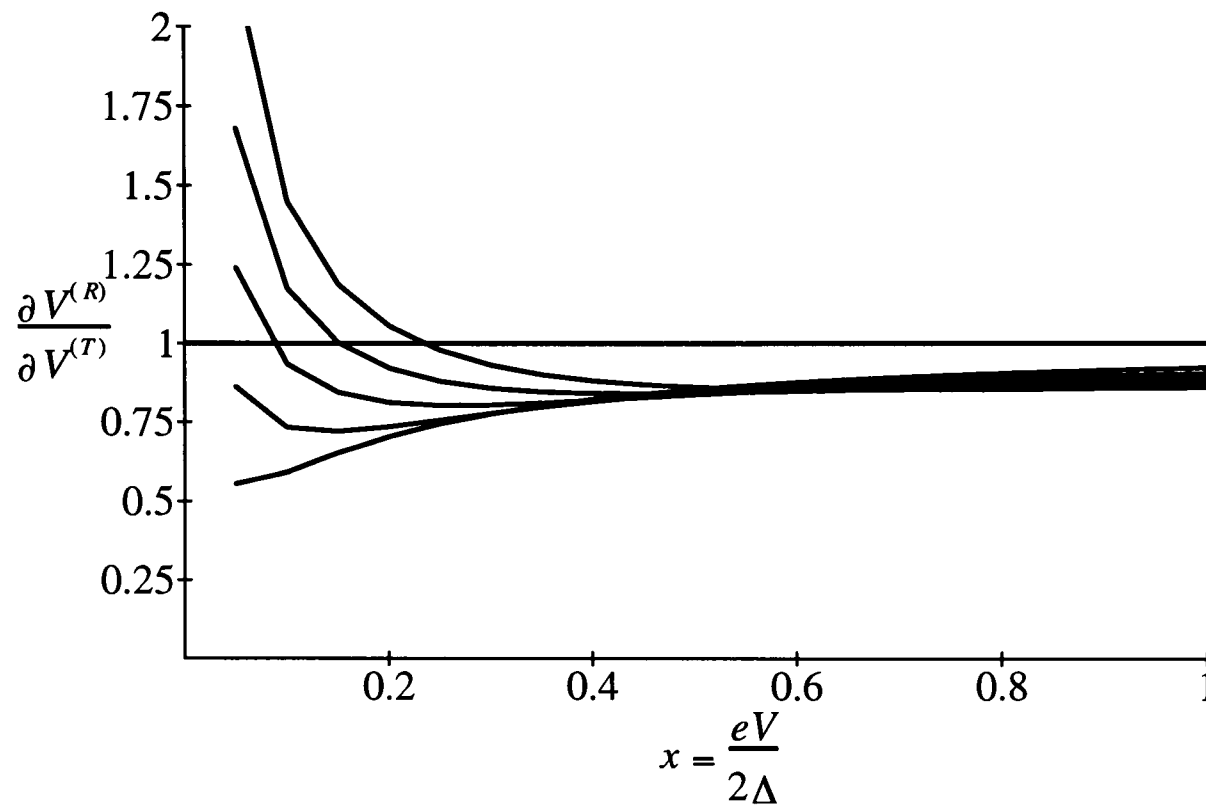


Figure 3.4.4 Ratio of voltage signals (for STJ at constant bias, injecting fixed number of quasiparticles) for Rd model divided by film temperature model. The top line is for $T/T_c=0.5$ running in equal steps down to 0.1. Horizontal scale is bias parameter x .⁴⁰

It is worth noting that the relationship between the voltage signals holds even in the presence of a temperature independent leakage current in the STJ of the form

$$I = I_{thermal}(x, y) + I_{leak}(x) \quad [3.4.27]$$

This can be represented by the substitution

$$f(x, y) \longrightarrow g(x, y) = \{f(x, y) + x\lambda(x)\} \quad [3.4.28]$$

In the case where the leakage current is caused by a small amounts of N-I-N tunnelling in parallel with the S-I-S characteristic of the junction the parameter λ is a constant and is given by the relationship:

$$R_{leak} = \frac{R_{nn}}{\lambda} \quad [3.4.29]$$

⁴⁰File:

It can be seen that the voltage ratio is unaffected since

$$\left. \frac{\partial g}{\partial y} \right|_x = \left. \frac{\partial f}{\partial y} \right|_x. \quad [3.4.30]$$

In conclusion it can be seen that using the R_D technique for calculating the excess numbers of quasiparticles does appear to be valid. It only breaks down when the change in the IV characteristic, near zero bias, associated with the increase in the number of quasiparticles within the junction becomes significant. It should be pointed out that the use of temperature of the junction in this section was done merely as a way to parameterise the change in IV applicable to the non-equilibrium increase in quasiparticle number and does not refer to the relationship between the quasi-thermal phonon distribution in either the superconducting films or substrate.

3.4.5. Sensitivity of STJ to Quasi-equilibrium Temperature Rise in Substrate and Film

The relationship introduced in Eq. 3.4.20 in the previous section, which relates the change in quasiparticle temperature of an STJ and its characteristic voltage at constant bias current, can be applied to the case of a quasi-thermal rise in the temperature of films and substrate together. Such a state occurs if there is no net transfer of phonons (in any frequency interval) between the film and substrate (on time scales longer than the recombination of quasiparticle in the film). This could be expected to occur when the phonon distributions in both the film and substrate become blackbody distributions at the same temperature.

We have shown already that the voltage change in the STJ at constant bias for a small excess of quasiparticles is given by

$$\delta V^{(R)} = \frac{1}{2e} \left(\frac{\delta n_{qp}}{N(0)} \right) \left(\frac{R_D}{R_{nn}} \right). \quad [3.4.31]$$

The change in quasiparticle number within the STJ is related to the junction change in temperature by

$$\frac{\partial n_{qp}}{\partial T} = \frac{\partial n_{qp}}{\partial y} \frac{\partial y}{\partial T} = 2N(0)(2\pi)^{\frac{1}{2}} k_B y^{\frac{3}{2}} e^{-y}. \quad [3.4.32]$$

Thus we can associate the equilibrium temperature rise of the crystal and film with a change in voltage bias by the relation,

$$\delta T = \frac{e}{(2\pi)^{\frac{1}{2}} k_B} \delta V^{(R)} y^{-\frac{3}{2}} e^y \left(\frac{R_D}{R_{nn}} \right)^{-1}. \quad [3.4.33]$$

As long as the experimentally determined R_D is used (and not the theoretical value) this relationship holds even in the presence of temperature independent leakage current.

In general, this expression remains valid as long as (i) δn_{qp} , the excess density of quasiparticles is small compared with n_{qp} , the thermal equilibrium density, and (ii) the heating of the junction due to the bias current is not significant.

3.4.6. Correspondence of Blackbody Phonon Spectrum and Quasiparticle Number in Superconductor

Given the analytic relationships derived in the previous section it is worth considering the underlying mechanism of detection of phonons, both sub and above gap, by an STJ.

Eisenmenger [60] pp110 discusses the sensitivity of STJs to phonons, $\Omega < 2\Delta$. He concludes that the ratio of the sensitivities for sub- to above-gap phonons is $< 10^{-4}$. This was verified experimentally by the failure to detect ballistic phonons, $\Omega \sim 400 \mu\text{eV}$, using Sn-Ox-Sn junctions [60, 66]. At higher temperatures, $T > 0.5T_c$, increased phonon absorption can result in finite low energy phonon signal contributions as was found with Al detectors [60, 66].

The reduced sensitivity to sub-gap phonons at low temperatures can be seen to arise from two mechanisms. (i) The strong temperature dependence of the ratio of normal to superconducting phonon absorption coefficients in the thin films and (ii) the rapid decay of quasiparticle excited states back to the gap edge which is the only possible way in which the quasiparticles in a superconductor can couple to sub-gap phonons. For the range $0 < \Omega < 2\Delta$ the BCS expression for low temperature ultrasonic attenuations is

$$\alpha_s = 2\alpha_N \left(e^{\frac{\Delta(T)}{k_B T}} + 1 \right)^{-1}. \quad [3.4.34]$$

For Sn STJs at 1 K we obtain

$$\alpha_s = 3 \times 10^{-3} \alpha_N, \quad \frac{\Delta}{k_B T} \approx 6.5 \quad [3.4.35]$$

To first order the sub-gap phonons do not give rise to any additional tunnelling current since the number of quasiparticles has not changed. However, they may cause variations in the tunnelling current due to the change value of the density of states, in the opposite STJ film, available to the excited quasiparticles. The opportunity for the quasiparticles to tunnel in the excited states will be short lived since they will rapidly relax back to the gap edge with times over two orders of magnitude shorter than the effective recombination time for typical STJ operating temperatures below about $0.5T_c$. This places an upper limit on possible effects due to these excited quasiparticles of 10^{-2} that of those at gap edge. The combination of these two effects leads to a maximum sensitivity to subgap phonons some four orders of magnitude lower than those above gap which will be detected directly by the STJ via pair breaking.

If we consider an STJ on substrate at times longer than the thermalisation time of the phonon signal within the substrate, then a component of this blackbody distribution will be above gap. Consequently an initially subgap phonon population will ultimately produce above gap phonons through the mechanism of thermalisation.

Considering the behaviour of a superconductor in contact with a blackbody source of phonons the processes affecting the quasiparticle population can be summarised:

Pair Breaking: $\Gamma_{pb}(\Omega, E \approx \Omega - \Delta) \rightarrow qp^\dagger(\Delta), qp^\dagger(\Omega - \Delta)$

Quasiparticle Recombination: $\Gamma_r(E_1 \approx \Delta, E_2 \approx \Delta) \rightarrow qp(\Delta), qp(\Delta)$

Quasiparticle Scattering (Phonon Emission): $\Gamma_s^{(Emission)}(E_1, E_2 \approx \Delta) \rightarrow qp^\dagger(\Delta), qp(E_1)$

Quasiparticle Scattering (Phonon Absorption): $\Gamma_s^{(Absorption)}(E_1 \approx \Delta, E_2) \rightarrow qp(\Delta), qp^\dagger(E_2)$

where $qp^\dagger(E)$ and $qp(E)$ represent the creation and annihilation of quasiparticles at an energy E . We have not included the change in the phonon states arising from these

processes since it is assumed that in thermal equilibrium the phonon states remain at the blackbody distribution. The energies of the quasiparticle states have been shown at their most probable values.

Of the four processes the last two conserve quasiparticle number and to first order do not effect the tunnelling current observed in a STJ biased well below gap edge (see argument in §3.4.5). In thermal equilibrium these two processes would be expected to balance.

The first two processes would also be expected to balance when the phonon and quasiparticle systems are in thermal equilibrium. The rate of creation of quasiparticles will be proportional to

$$\frac{\partial N_+^{(qp)}}{\partial t} \sim \Gamma_{pb} N_{\Omega \geq 2\Delta}^{(phonon)}. \quad [3.4.36]$$

The number of phonons above 2Δ in a blackbody spectrum is given by (assuming simple dispersion relation for phonons of three modes with the same velocity)

$$N_{\Omega \geq 2\Delta}^{(phonon)} \cong \frac{3}{2\pi^2} \frac{1}{(\hbar c)^3} \int_{2\Delta}^{\Omega_D} \frac{d\Omega \Omega^2}{e^{\frac{\Omega}{k_B T}} - 1}. \quad [3.4.37]$$

In the experimental situations we are considering $T \ll 2\Delta \ll \Omega_D$ and the integral can be approximated to have a temperature dependence (ignoring constants)

$$N_{\Omega \geq 2\Delta}^{(phonon)} \sim \int_{2\Delta}^{\infty} d\Omega \Omega^2 e^{-\frac{\Omega}{k_B T}} \sim T e^{-\frac{2\Delta}{k_B T}}, \quad T \ll 2\Delta \ll \Omega_D. \quad [3.4.38]$$

Considering the rate of annihilation of quasiparticles in the superconductor,

$$\frac{\partial N_-^{(qp)}}{\partial t} \sim \Gamma_r N^{(qp)}, \quad \Gamma_r \sim N^{(qp)}. \quad [3.4.39]$$

At thermal equilibrium the rates of these two processes balance implying a temperature dependence for $N^{(qp)}$ of the form

$$\frac{\partial N^{(qp)}}{\partial t} = \frac{\partial N_+^{(qp)}}{\partial t} - \frac{\partial N_-^{(qp)}}{\partial t} = 0 \Rightarrow \left(N^{(qp)}\right)^2 \sim T e^{-\frac{2\Delta}{k_B T}}, \quad [3.4.40]$$

$$N^{(qp)} \sim T^{\frac{1}{2}} e^{-\frac{\Delta}{k_B T}}.$$

[3.4.41]

Of course, this is precisely the form demanded by the statistics governing the quasiparticle gas. The analysis does, however, highlight that to first order the temperature sensitivity of the quasiparticle number in a superconductor can be related to the number of phonons in the superconductor above the superconducting gap.

3.4.7. Summary of Parameterisation of STJ Voltage Signals

Parameter	Value for STJ voltage change, δV ⁴¹
1. Rate of phonon energy entering junction ⁴² (Flux) $\delta\Phi$	$\frac{2\Delta}{2e\Gamma_{tun}R_D(T)\tau_r(T)}\delta V, \quad t \gg \tau_{RC}, \tau_r$
2. Quasiparticle Temperature Rise in STJ, δT	$\frac{e}{(2\pi)^{\frac{1}{2}}k_B}y^{-\frac{3}{2}}e^y\left(\frac{R_{nn}}{R_D}\right)\delta V$
3. Excess Quasiparticle Number in STJ, δN	$\frac{1}{e\Gamma_{tun}R_D}\delta V$

Table 3.4.4 Conversions from voltage signal to other parameterisations.

Table 3.4.4 gives a number of parameterisations of the change in signal voltage in a STJ. The first expressions relates to the rate at which energy is entering the STJ films. The second two expressions reparameterise the instantaneous voltage of the STJ in terms of either a quasiparticle excess or a nominal temperature rise in the films.

It can be seen that the temperature dependence of the conversion factors for the flux and temperature rise conversions are very different. The inverse of the STJ dynamic resistance, $(R_D)^{-1}$, is common to both and will increase as the temperature falls ($\sim e^{\Delta/k_B T}$ in an ideal STJ). The remaining temperature dependencies are

⁴¹ y is the dimensionless inverse temperature defined in Eq. 3.4.14.

⁴²For STJ voltage signals that correspond to phonon fluxes which are changing rapidly on the scale of τ_r we employed the Fourier deconvolution techniques outlined in §3.4.

$$\frac{\delta V}{\delta \Phi} \sim y^{\frac{1}{2}} e^y, \quad [3.4.42]$$

$$\frac{\delta V}{\delta T} \sim y^{\frac{3}{2}} e^{-y}, \quad [3.4.43]$$

where the dependence in Eq. 3.4.42 comes from Eq. 3.3.11 for τ_r . Figure 3.4.5 shows Eq. 3.4.42 and 3.4.43 for arbitrary normalisation, in a temperature range (given by y) for typical STJ operation.

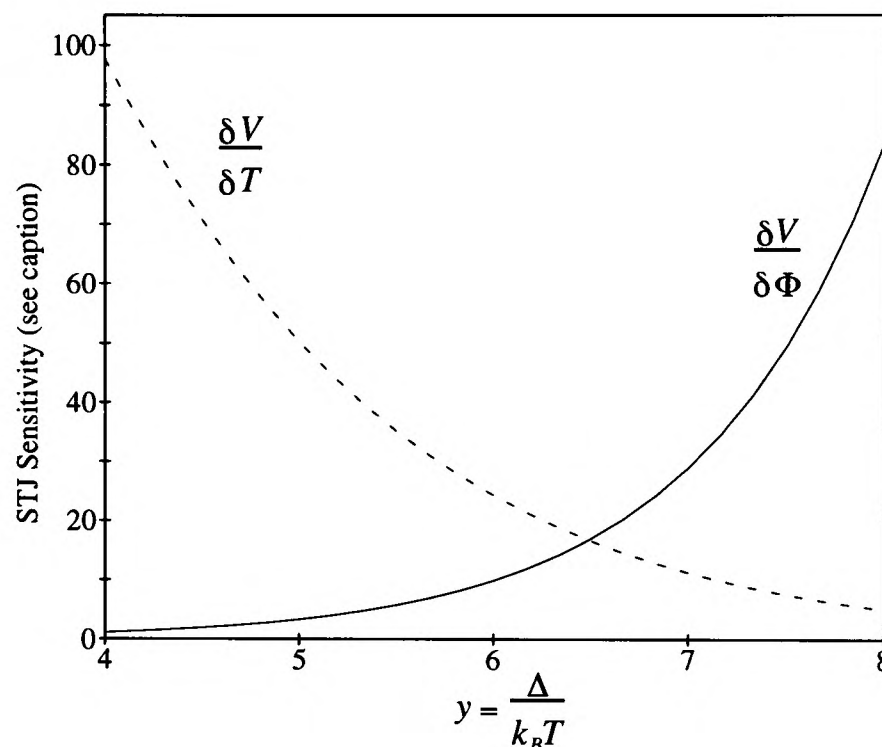


Figure 3.4.5 The relative sensitivity of the voltage signal, δV , from an STJ to a fixed above-gap phonon flux (line), $\delta \Phi$, and a temperature rise (dashed line), δT , plotted against reciprocal temperature. We have not included the temperature dependence of the device dynamic resistance which is the same in both cases.

It can be clearly seen that as the temperature of the STJ is lowered the sensitivity⁴³ of the STJ falls for a fixed temperature rise, whereas, it increases for a fixed phonon flux.

3.5. MODELLING DIFFUSION TRANSPORT IN NIOBIUM

3.5.1. Introduction

In order to model the diffusive transport of phonons and quasiparticles we investigated the solutions of the diffusion equation, using boundary conditions

⁴³We have not included the dynamic resistance temperature dependence since this is the same for both terms.

corresponding to the experimental crystal geometries, with a $\delta(\vec{x},t)$ -function source term. The simple diffusion equations is

$$\nabla_{\vec{x}}^2 \Psi = \frac{1}{D} \frac{\partial \Psi}{\partial t}, \quad [3.5.1]$$

where Ψ is the number density of the quantity diffusing, D is the diffusion coefficient and $\vec{x}$ and t are the space and time co-ordinates, respectively. Analytic solutions of Eq. 3.5.1 are possible, using Green's function techniques. However, they grow rapidly in complexity as the boundary conditions become more detailed. Greater flexibility is available if the diffusion equation is solved numerically. Arbitrary boundary conditions can be applied (for instance, a spatially varying absorption coefficient at the boundary) more easily.

We will briefly detail the salient points of our algorithm.

3.5.2. Program Algorithm

The simple numerical diffusion model is based on a time-step algorithm (which is derived by considering Eq. 3.5.1 on a discrete time and space lattice)

$$\Psi_n^{t+1} = \Psi_n^t + \delta \sum_{i=0,1,2} \left\{ \Psi_{(n-1)_i}^t + \Psi_{(n+1)_i}^t - 2\Psi_{n_i}^t \right\} \quad [3.5.2]$$

The t -index is the time step, and the n -index is the position of each spatial box in one of three dimensions (given by the sub-index i) and are shown in Fig. 3.7.1. The parameter

$$\delta = \frac{D}{\left(\frac{(\Delta x)^2}{\Delta t} \right)}, \quad (\delta < \frac{1}{2N}) \text{ where } N \text{ is the number of space dimensions,} \quad [3.5.3]$$

is the dimensionless diffusion parameter. N is the number of spatial dimensions used in the model (3). Δx and Δt are the model box size and time step and are chosen to meet the inequality in Eq. 3.5.3 ⁴⁴. For typical simulations of diffuse phonon or quasiparticle transport $\Delta x = 0.5 \text{ mm}$ and $\Delta t = 50 \text{ ns}$ will suffice. This represents a large number of individual calculations for a crystal of dimensions, $(10 \text{ mm})^3$, over a time of $100 \mu\text{s}$,

⁴⁴It can be shown that this algorithm becomes unstable if this inequality is not met.

however, current computer technology is well able to handle this. Intrinsically stable algorithms are available ⁴⁵ which do not require such small time steps, however, they introduce further complexity in determining the correct boundary conditions.

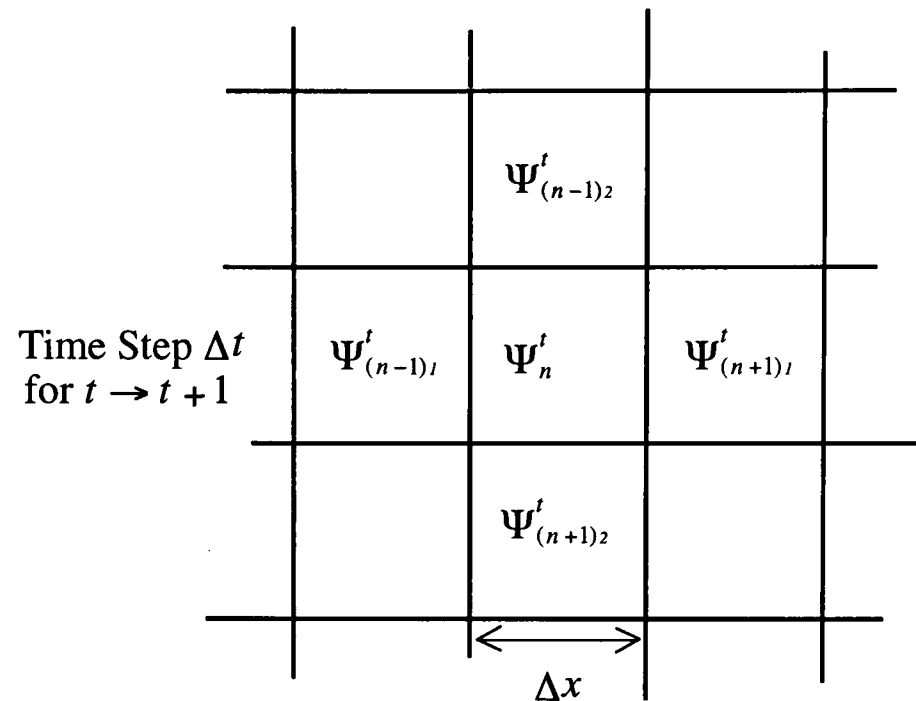


Figure 3.5.1 Two out of the three spatial dimensions are shown for the box diffusion simulation.

For the non-implicit solution technique we employed, the numerical imposition of the boundary conditions follows from the derivative form for the concentration gradient at the surface

$$\left. \frac{\partial \Psi}{\partial x} \right|_{\text{Perp.}} = -h \Psi, \quad [3.5.4]$$

where h is a parameter ($0 \leq h \leq 1$) determining the gradient of the concentration at the boundary. The derivative is evaluated for a direction, x , perpendicular and into the surface. The condition $h = 0$ corresponds to a fully reflecting surface. This parameter relates to the actual reflection coefficient, R , for the process being considered by

$$1 - R = h\lambda, \quad [3.5.5]$$

where λ is the m.f.p. of the process (with diffusion coefficient, D) such that (with a propagation velocity, c)

⁴⁵A review of the techniques available for the numerical solution of partial differential equations is available in Lapidus and Pinder or Press et al.[67]

$$D = \frac{1}{3} c \lambda \quad [3.5.6]$$

We considered improving the fit to the experimental phonon data by including a term in Eq. 3.5.1 which would reflect the finite propagation velocity of the particles diffusing ⁴⁶. One possible form is that of the “Telegraph Equation” [68],

$$\nabla_{\vec{x}}^2 T = \frac{1}{D} \frac{\partial T}{\partial t} + \frac{1}{c^2} \frac{\partial^2 T}{\partial t^2} \quad [3.5.7]$$

The equation for the development of the signal in the discrete model becomes

$$\Psi_n^{t+1} = \Psi_n^t + \left(\frac{\varepsilon \delta}{\varepsilon + \delta} \right) \sum_{i=0,1,2} \left\{ \Psi_{(n-1)_i}^t + \Psi_{(n+1)_i}^t - 2\Psi_{n_i}^t \right\} + \left(\frac{\delta}{\varepsilon + \delta} \right) \left\{ \Psi_n^t - \Psi_n^{t-1} \right\}. \quad [3.5.8]$$

The parameter

$$\varepsilon = \frac{c^2}{\left(\frac{\Delta x}{\Delta t} \right)^2}, \quad [3.5.9]$$

is the dimensionless ballistic parameter. The ratio of δ and ε is related to the dimensionless scattering time of the diffusion mechanism,

$$\frac{\delta}{\varepsilon} = \frac{\tau}{\Delta t}. \quad [3.5.10]$$

The replacement

$$\delta \rightarrow \frac{\varepsilon \delta}{\varepsilon + \delta}, \quad [3.5.11]$$

(comparing Eq. 3.5.2 and 3.5.8) reduces the diffusion term which is required so that the model does not predict the arrival of particles before their maximum speed of propagation. The behaviour of this expression in 3-D situations fails to account for the behaviour of ballistic component in the signal adequately ⁴⁷. The parameter, Ψ , cannot

⁴⁶In the limit that λ becomes significant compared with the source–detector distance the simple diffusion equation will predict the arrival of a significant part of the signal at a time before that estimated for ballistic propagation, at velocity c .

⁴⁷The shape of the leading edge of the signal (arising from a $\delta(t, \vec{r})$ -function source) is dispersed, rather than just attenuated, contrary to the required behaviour for the solution.

simultaneously parameterise the ballistic and diffuse component. This technique was not pursued further.

An alternative approach would be to consider solutions of the full form of the Boltzmann equation. This would allow us to account for the variation in phonon m.f.p. with energy which is apparent from the experimental results on phonons (see Chapter 5). We intend to perform such an analysis in the future.

3.5.3. Conclusion

The simple discrete diffusion model, given by Eq. 3.5.2, was used to analyse phonon and quasiparticle signal shapes when the m.f.p.'s were much less than the crystal dimensions. We also applied analysis based on the reflection coefficient, R , to get some feel for how the signal shapes may be altered for non-number conserving interactions at the surfaces of the crystal. A more complete analysis will be undertaken in the future using solutions of the energy dependent Boltzmann equation.

4. EXPERIMENTAL TECHNIQUES

Theory is a good thing but a good experiment lasts for ever.

Peter Leonidovich Kapitza

4.1. INTRODUCTION

The experiments detailed in this work were conducted on single crystals of Nb of two purities (RRR~10–60 and 10^4). In both cases the crystals were supplied as 100 mm boules (diameter 10 mm) which were cut into cylinder lengths of between 3–15 mm. A number of different types of superconducting tunnel junctions (STJs) were fabricated on the surface of the crystals. These devices were typically composed of thin films (1000–3000 Å) of Al, Sn, In and Pb. The manufacture of high quality STJs on the surface of the Nb posed a number of problems. We developed techniques for successful Nb surface preparation, manipulation of the crystals during thermal evaporation of the thin films, and for making electrical contact to the STJs prior to cooling to temperatures in the range 0.1–10 K. The bulk of the experiments (detailed in Chapters 5 and 7) were performed in a Cochise ^3He cryostat with a base temperature of 326 mK. Most of the proximity layer work, reported in Chapter 6, was performed in a ^4He cryostat, with a base temperature of 1.6 K. We also report an early result for particle detection (Chapter 7) in an Oxford

Instruments dilution refrigerator although this facility was being set up as this work was completed.

In the following sections we will detail a number of experimental techniques where they are important to the understanding of the devices and results.

4.2. NIOBIUM SINGLE CRYSTAL PREPARATION

The crystal samples were cut using spark erosion⁴⁸ from two Nb single crystal rods of diameter 10 mm.

The first boule was supplied by Metals Research Limited⁴⁹, had a nominal 5x9's purity, and was oriented in the [100] direction. Prior to sectioning the rod had been stored at room temperature in air. The samples were spark cut from the rod to 3 mm thicknesses (2g) and then the faces either mechanically polished using a diamond paste and silk pad, or spark cut again on a finer wheel (the latter causing less mechanical damage to the crystal surface). All subsequent manipulation of the samples and their mounting avoided any mechanical contact with the upper surface. An initial measurement of the RRR, by ourselves, using the eddy-current decay method gave a residual resistivity ratio at 10 K of ~10 (relative to the resistance at room temperature). A number of samples were outgassed for 3 hours at 850 °C at a pressure $< 10^{-7}$ torr. At this temperature it was calculated the hydrogen content would fall to less than 1 ppm. The original motivation for reducing the dissolved gas content of the crystal was to improve its RRR (~60), however, hydrogen outgassing became crucial to the successful manufacture of junctions directly on the surface of these crystals. Snow flake-like patterns grew on the surface of crystals which had not been outgassed when they were cooled below 1.5K. The structures were about 100 μm across (see Fig. 4.1) and so disrupted the thin films of the STJs which are 10 to 10,000 Å thick. The appearance of these features at such a low temperature was surprising and was probably due to the precipitation of hydrides. It was subsequently shown that the

⁴⁸A low voltage setting was recommended to avoid cracking of the oil, which introduces C into the Nb.

⁴⁹Cambridge, UK

mechanical polishing stage of surface preparation appeared to sponsor this effect and the effect was eliminated by using chemical rather than mechanical polishing.

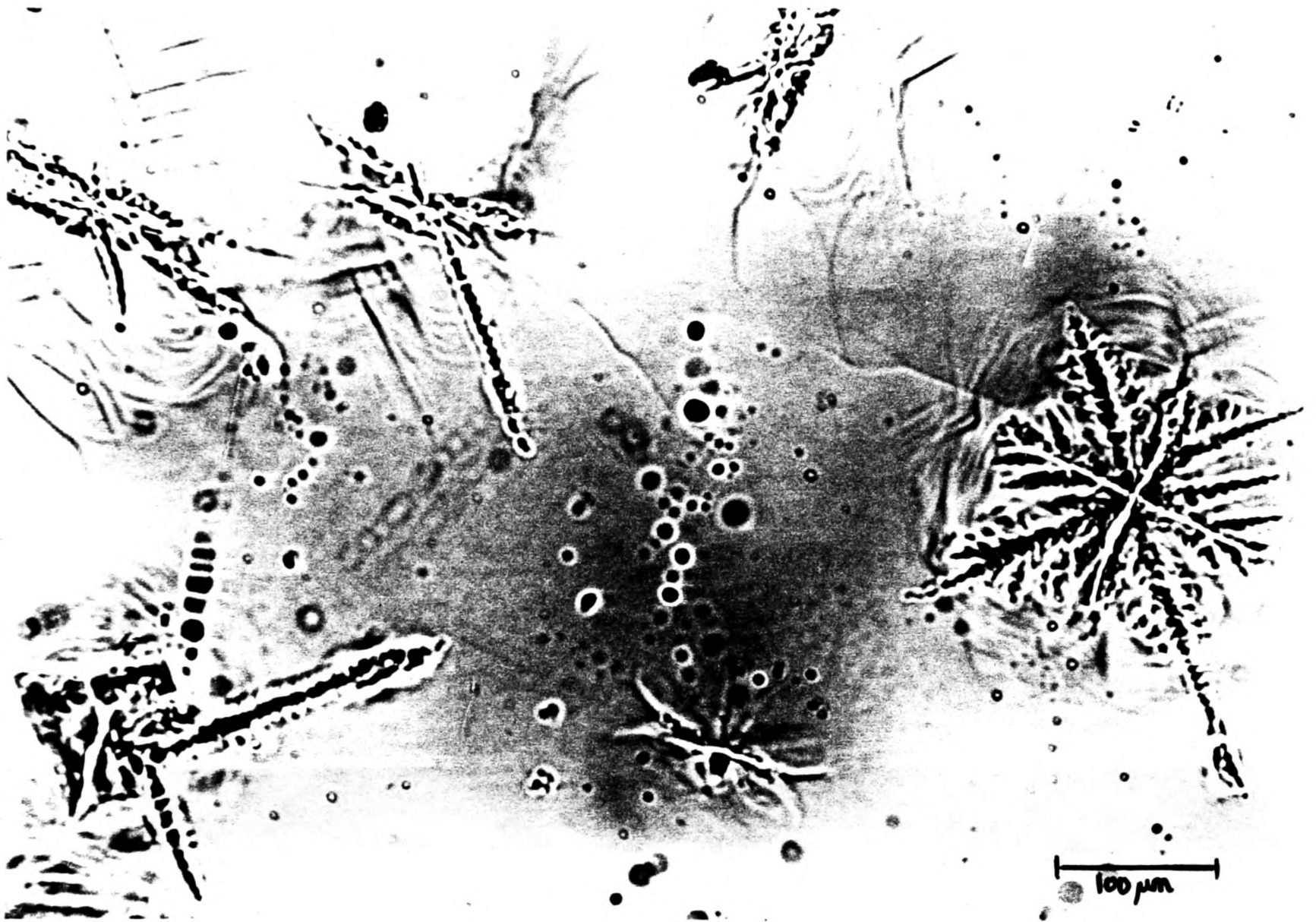


Figure 4.2.1. “Snow-flake” patterns that appeared on Nb surface when cooled below 1.5 K, due to mechanical polishing during surface preparation. The feature in the bottom right-hand corner is about 100 μm in width.

The second Nb boule was supplied by Dr Klaus Schulze⁵⁰. He estimated that the crystal RRR was $\sim 10^4$ with < 2 ppm of Ta and a total gas content ~ 5 ppm. The crystals were grown ([110] orientation) and refined using a multiple-pass vertical e-beam refining technique. The main purification process was the evaporation of impurities at a temperature of ~ 2500 °C and a pressure of 10^{-8} torr. The melting point of Nb is 2741 K.

⁵⁰Max-Planck Institute für Metallforschung, Stuttgart. Unfortunately Dr Schulze died during the course of this work. He was a leading expert on the refining and growth of ultra-high-purity single crystal Nb. [69, 70]

These crystals were initially spark cut into thicknesses of 5–15 mm. They were then spark cut again on a finer wheel to reduce the surface roughness. No mechanical polishing was used for reasons outlined in the previous paragraph.

The crystal surface was polished chemically to a mirror smooth appearance ⁵¹ prior to STJ fabrication. A standard Nb polish (1 conc. HF : 2 conc. HNO₃) was used for periods of 30 secs. to 5 mins. It was important to agitate the crystal to avoid gas build-up which would create irregularities in the Nb surface. Surface layers were removed at 10–50 $\mu\text{m min}^{-1}$ depending on the relative concentration of the two acids. The crystals were subsequently washed in high purity de-ionised water. It was noted that pitting of the crystal surface could occur if the acids were allowed to warm above room temperature or the proportion of conc. HF was above 50%. After chemical polishing the crystals were held in specially constructed clamps for mounting in the thermal evaporator.

Recycling of crystals which had STJ devices on their surface could be achieved using chemicals that removed the SiO ⁵² and metal films (used for STJ fabrication) without etching the Nb which was then chemically repolished.

4.3. DEVICE FABRICATION

4.3.1. Introduction

STJs were fabricated on the surface of the Nb crystals using standard metal evaporation techniques. The junctions fell broadly into three categories: (i) Nb_{crystal}–SiO–X–Ox–X ⁵³ junctions formed by two thermally evaporated metal films, insulated from the Nb crystal surface by a 5000 Å layer of SiO ⁵⁴; (ii) Nb_{crystal}–NbOx–X junctions, with the

⁵¹No features were visible on a scanning electron microscope with a resolution ~ 100 Å.

⁵²Layers of SiO were successfully removed from Nb crystal surfaces using 10–12 molar aqueous NH₄F. The solution can be buffered with NH₄OH to yield a pH of 9 as suggested in Massel and Glang §7-38[71] and Vossen and Kern p413[72] although omitting the latter step avoided the formation of precipitates on the surface of the Nb crystal. The solution was raised to 80 °C giving an etch rate of ~ 2000 Å min⁻¹.

⁵³“X” represents a thin metal superconducting film such as Al, Sn, In or Pb.

⁵⁴The quality of the SiO films was important for the successful fabrication of the STJs. SiO films were deposited at rates ~ 20 Å s⁻¹ at pressures below 5×10^{-6} torr. This regime was achieved by outgassing of the SiO and Ta boat prior to beginning fabrication.

crystal as one electrode, using the Nb native oxide as the tunneling barrier and thermally evaporating a second film; and (iii) Nb_{crystal}-Al-AlO_x-X junctions similar to (ii) except that a layer of Al was deposited directly on the surface of the Nb crystal which was then oxidised to form the tunneling barrier. These three geometries are shown schematically in Fig. 4.3.1. SiO was used to define the junction areas in the case of STJs (ii) and (iii).

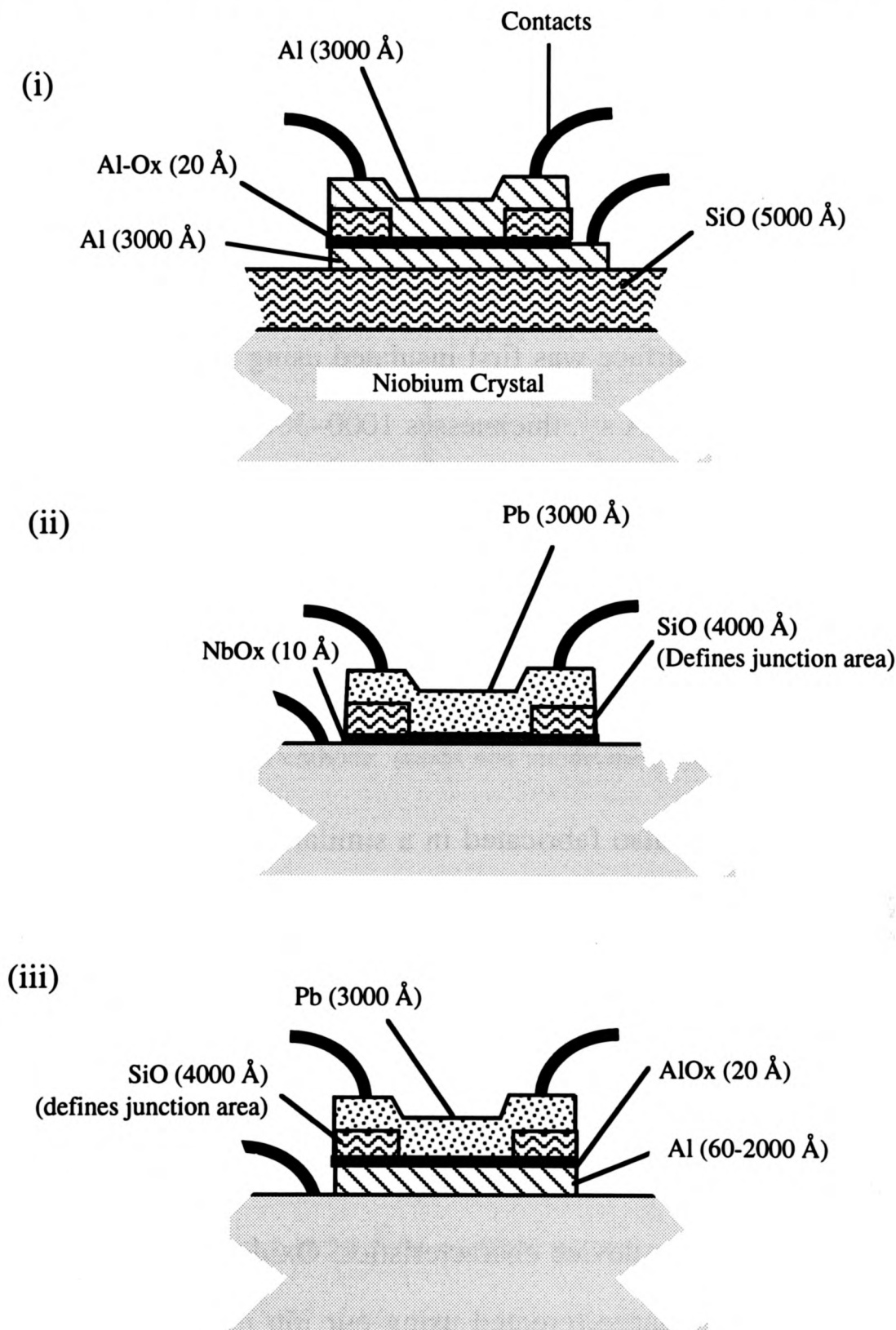


Figure 4.3.1. STJ geometries, described in the text, on the surface of a Nb crystal
(i) Nb_{crystal}-SiO-Al-Ox-Al (ii) Nb_{crystal}-NbOx-Pb (iii) Nb_{crystal}-Al-AlOx-Pb.

The thermal evaporation system had provision for 12 shadow masks and 6 evaporation sources. The masks and sample are independently rotating to be positioned

over any evaporation source position. An ion mill⁵⁵ was available for cleaning the crystal surface prior to film deposition. Film thickness measurement used an Edwards FTM4 6 MHz oscillator.

The vacuum is provided by a 300 l s⁻¹ turbo-molecular pump and a 600 l s⁻¹ cryopump which was mounted directly below the evaporation chamber. The typical base pressure was 10⁻⁶ torr.

4.3.2. Al-Ox-Al and Sn-Ox-Sn Superconducting Tunnel Junctions Insulated from the Crystal Surface

Al-Ox-Al STJs were fabricated on the surface of the Nb using a simple cross strip geometry. The Nb crystal surface was first insulated using 5000 Å of SiO. The Al films were deposited at a rate of 20 Å s⁻¹, thicknesses 1000–3000 Å. Oxidation of the first film was made immediately after deposition at an O₂ pressure of 1 torr for 3 mins⁵⁶. The second film was deposited, after oxidation, when the evaporator had been pumped to a pressure of better than 5 × 10⁻⁶ torr. The junction was then covered in a second SiO film of thickness 1 µm to prevent deterioration of the junction quality when exposed to the atmosphere.

Sn-Ox-Sn STJs were also fabricated in a similar pattern to that of the Al-Ox-Al STJs. However, junctions made on an evaporated SiO layer (on either Si or Nb substrates) showed significant leakage currents. We were able, however, to make good Sn STJs on a 3000 Å oxide layer grown on the Nb surface using wet anodisation. The technique is outlined in a following paragraph. The Sn films were deposited (at rates of 80–100 Å s⁻¹) in a 10⁻⁵ torr background of O₂. These conditions are known to reduce the Sn crystal size in the films and improves the device characteristics. Oxidation of the first Sn layer was achieved using a glow discharge (created using our ion mill at 2 kV potential) with a continuous flow of O₂ at 12 mtorr for 4 minutes. The reliability of Sn STJs was significantly inferior to those of the made from Al. We would hope in the future to

⁵⁵Ion Tech Model FAB 100W. Ar gas was used at a pressure ~1 mtorr.

⁵⁶This results in typical junction resistances of 2 × 10⁻⁵ Ωcm².

substitute Ta, since it has similar superconducting properties to Sn, but is known to form a more robust oxide.

The Nb crystal surface could be anodised to form a native oxide of over 1000 Å thickness which appeared to be robust enough to fabricate junctions on. The experimental arrangement used is shown in Fig. 4.3.2. The anodisation solution was made up from Ammonium Pentaborate : Ethylene Glycol : Water in the ratio 156 g : 1120 ml : 760 ml. A few drops of the anodisation solution were put on the surface of the crystal so that the meniscus was covering the entire top surface but not overflowing the edges. A Platinum wire was used as the cathode (–ve) in the solution and a second anode wire (+ve) was pressed against the side of the Nb crystal.

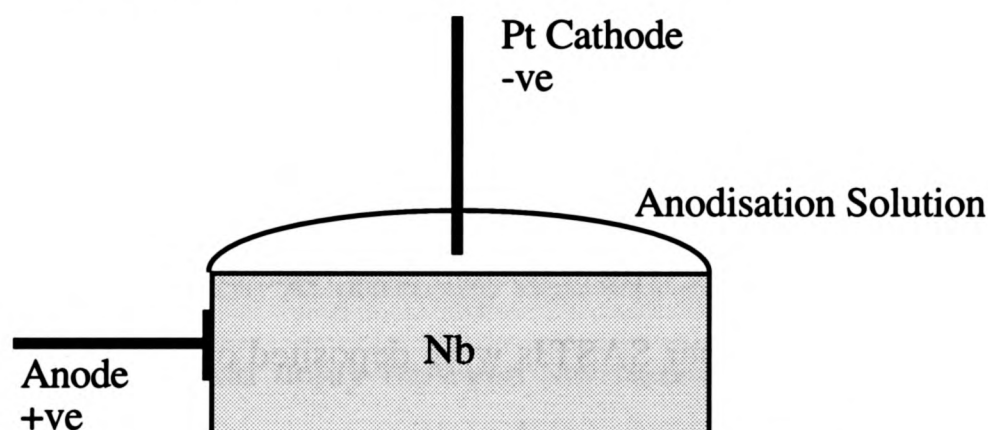


Figure 4.3.2. Arrangement of cathode, anode and anodisation solution for oxidation of Nb crystal surface.

The anodisation was begun with a current of 400 μA . The voltage drop across the cathode–anode at this current was ramped from 0–50 V over a period of 6 minutes. Subsequently a voltage limit insured that the voltage remained at 50 V while the current started decreasing. The process was stopped when the current reduced to 20 μA . This second stage lasted 5 minutes and insures that any holes in the oxide layer fill in.

The Nb_2O_5 layer is formed at 23 Å/V so 50 V produces an oxide of ≈ 1200 Å (which is yellow/green in colour). 8.5 Å/V of Nb are used in the oxide formation. Voltages above 60 V cause break down in the Nb oxide.

4.3.3. Superconducting Series Arrays of Tunnel Junctions

The techniques employed to make a single Al–Ox–Al STJ were extended to make a (24–400) series connected chains of Al STJs (SASTJ). These devices were used extensively in the particle detection work reported in Chapter 7. They have the advantage of covering a much larger area of the Nb crystal surface while maintaining the electronic time constant of a single STJ (see §5.5.7). The SASTJ was fabricated using just two separate Al depositions by arranging the shadow masks such that each island of Al from the first deposition overlaps with two islands from the second deposition. This approach has the added advantage that the oxidation condition for all the junctions is identical. The shadow mask technique is able to produce minimum island sizes of $\sim 200 \mu\text{m} \times 100 \mu\text{m}$ which permits a maximum junction density of $\sim 3000 \text{ cm}^{-2}$. We are still investigating the optimal number. The experiments reported in this work used individual SASTJs containing 150 junctions covering an area of $3 \times 6 \text{ mm}$ (with about 30% of this area actually covered in Al film). The typical normal state resistance for such a series is 3.5Ω . Devices were made in which four SASTJs were deposited on both faces of a Nb crystal. This allowed the simultaneous energy and position measurement of particle interactions (see Chapter 7 and photograph therein). The fabrication of this geometry required particular care in handling the crystals.

4.3.4. Superconducting Tunnel Junctions Directly on the Niobium Surface

In order to make STJs with the Nb crystal as one electrode, using the native oxide as the barrier, it was necessary to ion mill the surface of the crystal (to remove any oxide formed after the crystal cleaning) and to reoxidise the surface using analysis grade O_2 introduced into the chamber at 10 torr for 30 mins. It was demonstrated that omitting the milling step led to junctions that showed semi-conductor like properties⁵⁷ at low temperatures. Nb_{crystal}–Ox–Pb junctions that were made, following milling of the Nb

⁵⁷The junction resistance would increase by ~ 100 when cooling for 300–10 K. When the films were superconducting the junction IV would show μ -bridge characteristics at biases of a few meV and a steady fall in resistance as the bias was increased $\sim 100 \text{ mV}$.

surface, showed structure in the IV characteristics (see Fig. 6.2.1) at voltage biases of 2.8 meV which was attributed to damage during the milling operation. The minimum operating voltage of the ion mill in the evaporation system was 2 keV so we were unable to investigate the reduction of the damage layer by reducing the milling voltages. In addition, the IV characteristics showed the onset of an additional current at 1.3 meV which appeared to be caused by normal metal regions on the surface of the Nb. Inadequate milling was shown to increase the size of this leakage current, however, we were unable to reduce it below the contribution shown in Fig. 6.2.1. The leakage current is discussed further in §6.2.

We also fabricated Nb_{crystal}-Al-Ox-Pb STJs formed by depositing an Al layer directly on the Nb crystal, oxidising the Al, and then depositing the Pb probe film. The initial results (see §6.3.2) for Al proximity layers fabricated in the thermal evaporation system were disappointing. The apparent transmission probability of quasiparticles between the Nb crystal and the Al proximity film was extremely low (6×10^{-3}). During device fabrication, the typical delay between ion mill cleaning of the crystal and the deposition of the Al layer was 90 secs. The partial pressure of O₂ ($\sim 2 \times 10^{-7}$ torr) in this system was such that this delay considerably exceeds the mono-layer time. We believe that partial formation of a Nb native oxide barrier was disrupting quasiparticle transmission.

Significant improvement was achieved by moving the Al proximity layer fabrication into a sputtering system with a significantly better vacuum. The CVC DC magnetron sputtering system ⁵⁸ had a base pressure of 5×10^{-8} torr (after a 12 hour bake-out and 12 hours cooling) with typical oxygen and water partial pressures of 2×10^{-10} and 5×10^{-9} torr, respectively. Al films of 60–2000 Å were sputtered onto the Nb crystal, after ion mill cleaning at a succession of lower acceleration voltages (from 2 keV to 0.5 keV in steps of 0.5 keV). Unfortunately, there was no way of continuing the junction fabrication in this system and so the crystal was removed and placed quickly in the evaporation system. The transmission coefficient of quasiparticles at the Nb–Al interface

⁵⁸Situated in the thin film clean room facility at this laboratory

in these devices was found to be ~ 0.3 . The normal state resistances of the STJs were very high (due to the exposure of the clean Al layer to the atmosphere). This prevented their use for measurements of non-equilibrium signals in the Nb. We are hopeful that this composite fabrication technique will yield more useful STJs shortly.

4.3.4. Crystal Mounting and Electrical Contact to Thin Films

Following removal from the evaporator the Nb crystals were mounted in two piece OFHC Cu clamps. The base plate of the clamp could be screwed directly onto the sample stages of the cryostats. The clamps have a cylindrical internal profile which approximately matches the circumference of the Nb crystals. Cigarette paper soaked in GE varnish was used between the crystal and clamp to provide electrical isolation. The cross-sections of the Nb crystals deviated from perfect circles and so it was impossible to insure continuous contact around the edge of the crystal. GE varnish was used to take up some of the slop (≤ 0.5 mm). It was shown (see §5.6) that the thermal time constant of this arrangement was 15 ms for a 12 g Nb crystal. For the experiments performed this was adequate.

Electrical contact to the thin films on the Nb surface was achieved by using Cu foil strips ($50\text{ }\mu\text{m} \times 1\text{ mm} \times 10\text{ mm}$), soldered at one end to Cu-Capton contact strips glued to the clamp stage. The Cu foil was folded gently onto contact pads ($\sim 2\text{ mm} \times 2\text{ mm}$) which formed part of the thin films. Extreme care was required not to apply pressure to the Nb surface since this would rupture the SiO insulation rendering the device useless. In early experiments silver conducting paint ⁵⁹ was used to fix the Cu foil to the film contact pads. This proved too unreliable at low temperatures. Silver loaded epoxy ⁶⁰ was substituted with much more reliable results. Significant improvement of the contact adherence was also achieved by using small spots of In, deposited in the evaporator during STJ fabrication, beneath the contact pads. The pads were reinforced by insuring the SiO layer covering the STJ also partially covered the contact pads, as shown in Fig. 4.3.3.

⁵⁹RS Components, UK.

⁶⁰EPO-TEK 410 E, Promatech Ltd, Ashton Keynes, Wiltshire, UK.

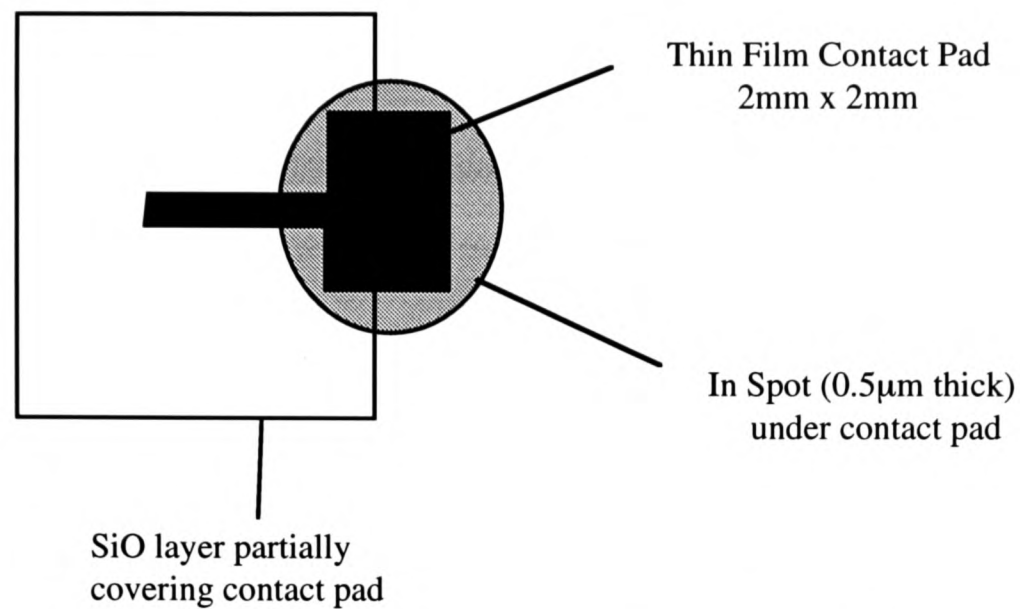


Figure 4.3.3 Structure of thin film contact pads to improve contact adherence to the SiO on the Nb crystal surface.

4.4. CRYOGENICS

4.4.1. He-3 Cryostat

The phonon and quasiparticle propagation experiments were conducted in a commercial ^3He cryostat (Cochise Instruments). This has a base temperature of 326 mK and a hold time of 6 hours (in the absence of additional heating of the stage). The samples (Nb crystal held in Cu clamps, described in §4.3.3) were mounted in vacuum on a copper sample stage in contact with the ^3He cold finger. Four optical fibres allowed laser pulses to be directed at the samples. Generally one fibre was used directly above the STJ on the crystal surface and a second one arranged symmetrically on the far face of the crystal.

The temperature was controlled using a small non-inductive resistance heater mounted on the back of the 350 mK plate. A Ge resistance thermometer ⁶¹ was fixed to the sample stage. The resistance was measured using an Oxford Instruments low current four terminal ohmmeter. The measuring current was 1 μA at 360 mK. Errors in measured temperatures are estimated to be within 2 mK in the region 0.33 K to 2 K. The thermometer calibration was obtained from a second calibrated GR200A. Between 2 K

⁶¹Lake Shore Cryogenics GR200A-100

and 4 K the temperature could be controlled by directly pumping on the ^4He bath. However, since the ^3He gas provides most of the thermal coupling between the sample on the cold finger and the ^4He reservoir temperature stabilisation in this region was difficult leading to greater uncertainty in the temperature readings.

4.5.2. Laser Pulses

The laser used was a small 5 W GaAs diode laser which emits at a peak wavelength of 9040 Å. The diode was driven at up to 45% of its maximum light output and the light was projected into a 200 µm-quartz optical fibre. The estimated collection efficiency of this arrangement was 10% (due to solid angle effects). Approximately 5 m of fibre was used to get the light down to the ^3He stage which has negligible attenuation. The maximum laser pulse width was 200 ns. We calculate that the energy per pulse is 50 nJ (300 GeV). This was confirmed using a calibrated light sensitive diode. A 95% reflectivity at the Nb crystal surface would lead to a pulse deposition energy of ~15 GeV.

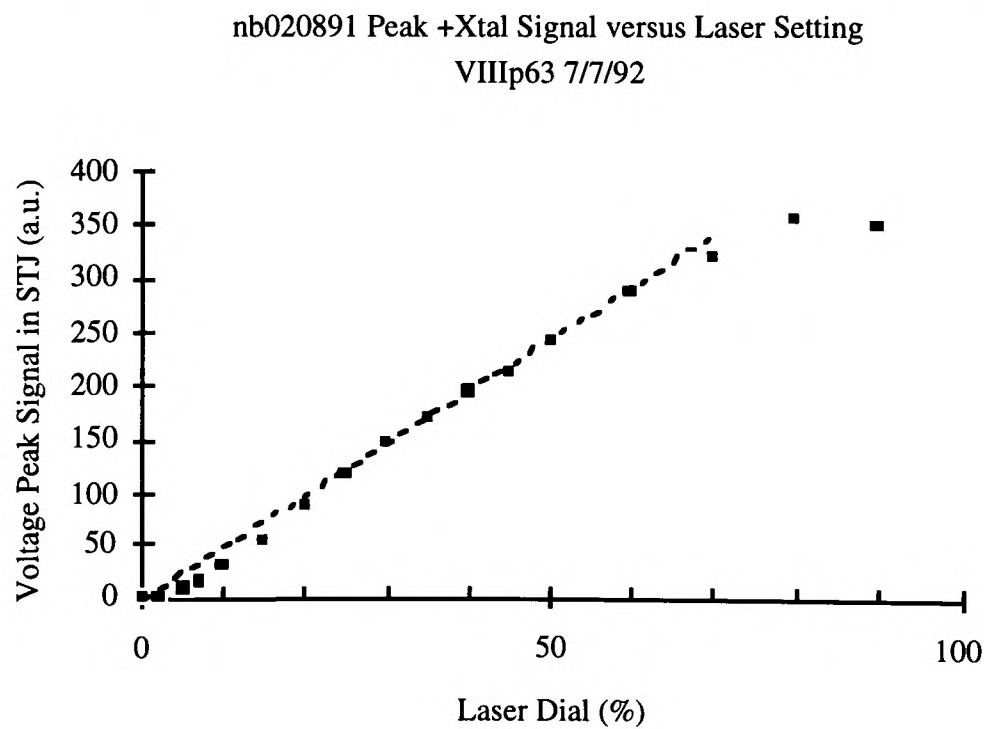


Figure 4.4.1 Energy of laser pulse with front dial setting of the laser power supply. An STJ was used to monitor the signal size. This trend is identical to that observed for pulse detection using a light sensitive diode.⁶²

⁶²VIIIp64

The laser power supply also includes an LED to provide an electrically isolated trigger pulse for signal averaging. Figure 4.4.1 shows a calibration of the laser against the current drive amplitude (as % of maximum dial setting).

4.5. ELECTRONICS

4.5.1. Laser Detection Experiments

For pulse amplification four FET parallel input amplifiers were used, mounted externally on the cryostat. The input to the pulse amplifier is also connected to a dc instrumentation amplifier to allow measurements of bias conditions. All the low-level electronics are powered using $\pm 12\text{V}$ battery supply to minimise mains pick-up.

The pulse amplifier (that was used for recording particle detection data and ballistic phonon signals) has a rise time of 20 ns and is ac coupled with three differentiating time constants (approximate net time constant $810\text{ }\mu\text{s}$ ⁶³). The amplifier noise is 0.35 nV/ $\sqrt{\text{Hz}}$ at frequencies in the range 0.01–1 MHz. Where appropriate the signals were also further amplified and put through a timing amplifier with integration time constants in the range 100 ns–1 μs to improve the signal to noise. The pulses were digitised using either a LeCroy 9400 125 MHz (8 bit/11 bit on repeat averaged signals) or a LeCroy 9314 300 MHz. The LeCroy scopes could be used to average repeated pulses from the laser excitation. The pulse data files were down-loaded to an IBM-PC via a GPIB connection. The were then transferred to a VAX cluster for analysis and display. The pulse files were corrected for the ac time constants of the amplifiers and could also be deconvoluted with the measured response of the STJ for direct illumination (see §3.6.1) to yield a pulse corresponding to the direct phonon flux into the junction.

⁶³ CR differentiating circuits were present in head the amplifier with time constant of 1.2 ms, 3 ms and 15 ms. During digital post-processing of the signals, where necessary, the time constants were folded back out of the data. Unless otherwise specified the pulse data shown in this work has been adjusted in this way.

4.5.2. Particle Detection Data Acquisition

The particle detection data shown in Chapter 7 was recorded using two separate techniques. During initial investigations we used an integrating amplifier, after the head amplifier (see §4.5.1), to integrate the voltage signal over a times of 10-200 μ s. The time was adjusted to give the maximum signal to noise ratio. The integrated pulses were recorded individually in the LeCroy scope using the scope threshold trigger to start pulse capture. After recording a single pulse it was transferred automatically to the IBM-PC which measured the pulse height relative to the base line (immediately proceeding the pulse start). The pulse was then discarded and the system was ready to record a new pulse. This system had a maximum acquisition rate of 3 Hz. This system was superseded by an off-line analysis which could be used once it was established that a particular detector geometry was working. The voltage pulses, direct from the head amplifier, were digitised in the LeCroy scope. 32 pulses were stored sequentially and then down-loaded to the IBM-PC which filed them to disk. This system was able to handle up to 2000 pulse events (8 Hz maximum) in a single run. The entire pulse series was down loaded to the VAX cluster and various fitting algorithms were applied to measure the pulse areas, rise times and decay times. These systems have now been replaced by a much faster dedicated pulse acquisition system which will run at 200 Hz and is under development at present.

4.5.3. Superconducting Tunnel Junction Current-Voltage Characteristics

For dc measurement of current voltage characteristics (IV) and derivatives a number of purpose built electronics units were used ⁶⁴. The IV measurements were made using standard instrumentation amplifiers to achieve low errors in gains and high input impedances. Derivatives were measured using a simple lock-in amplifier system ⁶⁵.

⁶⁴Designed by D.J.Goldie of this laboratory.

⁶⁵Analogue Devices 630AD balanced modulator/demodulator.

5. INVESTIGATION OF TRANSPORT PHENOMENA IN NIOBIUM

Concepts without factual content are empty; sense data without concepts are blind... . The understanding cannot see. The senses cannot think. By their union only can knowledge be produced.

Immanuel Kant

5.1. INTRODUCTION

The aim of this work was to investigate the feasibility of manufacturing low temperature particle detectors from single crystal Nb. The argument for using Nb in a dark matter search were presented in Chapter 2. However, the experimental requirements of building such a detector necessitate a detailed understanding of the behaviour of non-equilibrium phonon and quasiparticle signals in Nb. In the course of our investigations a number of difficult technical and experimental challenges were raised. Some were overcome and others remain. The successful fabrication of superconducting thin film devices on the surface of Nb was a pains-taking operation. The results reported in this Chapter and Chapters 6 and 7 represent only a small – but successful – fraction of the work carried out.

Relatively little is known about the non-equilibrium dynamics of phonon and quasiparticle propagation in bulk single crystal Nb. Existing studies have concentrated on

the measurement of the specific heat [73-79] and thermal conductivity [80-84] of bulk Nb: properties characterising the behaviour of near-equilibrium distributions of the phonons and electrons. Measurements of the propagation of phonons using ultrasonic techniques have also been made [35, 37, 85-88].

Nb is now in common usage in the manufacture of thin film superconducting devices. This work, however, is still in its infancy as far as the detailed understanding of quasiparticle dynamics in Nb. In principle, the use of bulk single crystal Nb should provide a simple system in which to study such behaviour. However, our investigations in this area have been plagued by the complexity of the surface physics of Nb (see the comprehensive summary by Halbritter [89]). Similar problems have hampered the development of the Nb thin film devices and have also been the major barrier to previous investigations of superconducting tunnel junction fabrication directly on Nb single crystals (see Chapter 6 and also [90] and references therein).

In the following sections we will discuss results of the observation of phonon and quasiparticle propagation obtained from single crystal Nb. The non-equilibrium signals were created using laser excitations and Joule heating of the Nb surface. Phonons in the Nb were detected using superconducting tunnel junctions (STJ) fabricated on the surface of Nb crystals. A comprehensive review of the techniques available can be found in Eisenmenger [60]. In summary, an STJ is formed by a superconductor-insulator-superconductor sandwich. The current in a STJ, biased at a voltage below twice the superconducting energy gap, depends on the number of quasiparticles in the films available to tunnel across the insulator. At temperatures significantly below the superconducting transition temperature, T_c , of the superconducting films the number of thermally excited quasiparticles becomes extremely small. If a high-energy phonon, Ω , is transmitted from the substrate (on which the STJ is made) into the superconducting thin film it will break a Cooper Pair in the film generating two quasiparticles. These quasiparticles will be available to tunnel through the S-I-S sandwich and will register as an increase in the tunneling current. The threshold for a phonon to be able to break a Cooper Pair is $\Omega \geq 2\Delta$ where Δ is the superconducting energy gap of the thin film. In our

experiments on Nb we have, for the most part, used STJs fabricated from Al and Sn which have phonon detection thresholds of $\sim 400 \mu\text{eV}$ and $1200 \mu\text{eV}$, respectively.

Phonon propagation in bulk superconducting Nb is limited by an upper energy, determined by $2\Delta_{\text{Nb}} \sim 3 \text{ meV}$ (see §3.3.6). Phonons above this threshold will rapidly break Cooper Pairs in the Nb itself creating quasiparticles (above-gap phonon m.f.p. $\sim 100 \text{ \AA}$). At temperatures significantly below the T_c of Nb, phonons at energies below the superconducting energy gap will propagate freely, unhindered by scattering from electrons. Given that Nb has the largest energy gap of the elemental superconductors its phonon propagation properties at low temperatures are more akin to those of dielectric crystals.

STJ junctions fabricated with the Nb crystal as one of the superconductors of the S–I–S sandwich were also shown to be sensitive to quasiparticle signals in the Nb. The quality of such junctions was found to be extremely poor, however. Difficulties in providing a clean interface at the Nb surface hampered work to demonstrate the effect of quasiparticle trapping (see §5.4). In the late stages of this research we developed techniques which demonstrate a significant improvement in the Nb interface and we hope to readdress the issue of trapping shortly.

Initial experiments were performed on low purity Nb crystals ($\text{RRR} \sim 10\text{--}60$). Propagation of both phonons and quasiparticles were found to be impurity limited (m.f.p.'s of $100 \mu\text{m}$ and $1000 \text{ \AA}$, respectively). Later experiments involved higher purity samples ($\text{RRR} \sim 10^4$) in which low energy phonons ($\Omega \sim 400 \mu\text{eV}$) were shown to propagate ballistically with m.f.p.'s in excess of the dimensions of the crystals studied ($5\text{--}15 \text{ mm}$). The quasiparticle m.f.p. also increased to $10 \mu\text{m}$. Unfortunately, this means that over distances between $2\text{--}10 \text{ mm}$ the phonon and quasiparticle signals tend to overlap. As a consequence ambiguities in the interpretation of some of our data arose.

The discussion in the rest of this chapter will go as follows. Firstly, we will provide evidence of the ballistic transport of phonons in high purity Nb and make an analysis of their propagation velocities based on the second-order elastic constants. This will include

a discussion of phonon focusing. We will then discuss results of the temperature dependence of the phonon m.f.p. at temperatures above 2 K arising due to scattering from thermally generated quasiparticles. In §5.4 we present results on the propagation of quasiparticles in low and high purity Nb samples. In §5.5 we discuss results that appear to indicate anomalously short thermalisation times for non-equilibrium phonon distributions in Nb below 2 K. These results have some important ramifications for the use of STJs on Nb for particle detection (see Chapter 7). We will also discuss a quantitative analysis of the signal sizes, which again is important from the point of establishing the energy detection threshold available in a Nb/STJ based particle detector. Our extensive analysis of the quasi-thermal tail observed in the STJ signals (at time-scales $\gg 100 \mu\text{s}$) is a consequence of some initial confusion we had over whether this was evidence of a quasiparticle signal in the Nb. It is clear that if we are to separate the evolution of the quasiparticle and phonon signals in Nb we must have a very detailed knowledge of the way they behave. We also introduce the series array of superconducting tunnel junctions (SASTJ). The SASTJs are used extensively in our work on detection of particle interactions in Nb. §5.6 outlines an independent verification of the thermal time constant of a Nb sample. The final section discusses, in some detail, the influence of different modes of excitation of the Nb which alters the non-equilibrium signal characteristics.

5.2. BALLISTIC PHONON PROPAGATION

5.2.1. Experimental Observations of Ballistic Phonons

The leading edge of non-equilibrium signals traversing the highest purity Nb crystals, generated by a laser excitation at the surface and detected by an Al STJ or SASTJ on an opposite face, were characterised by the ballistic propagation of phonons. Along the [110] axis (which was the major axis of the cylinder of all the high purity, $\text{RRR} \sim 10^4$, Nb samples) the phonons will propagate in three non-degenerate modes L (longitudinal), FT (fast transverse) and ST (slow transverse). In the lower purity

($RRR \leq 60$) Nb single crystals ballistic phonon propagation was almost absent over distances of a few mm. This was due to the much smaller phonon mean free path in these samples which was limited by impurity scattering. The observation of phonon diffusion in these samples is discussed in more detail in the next section.

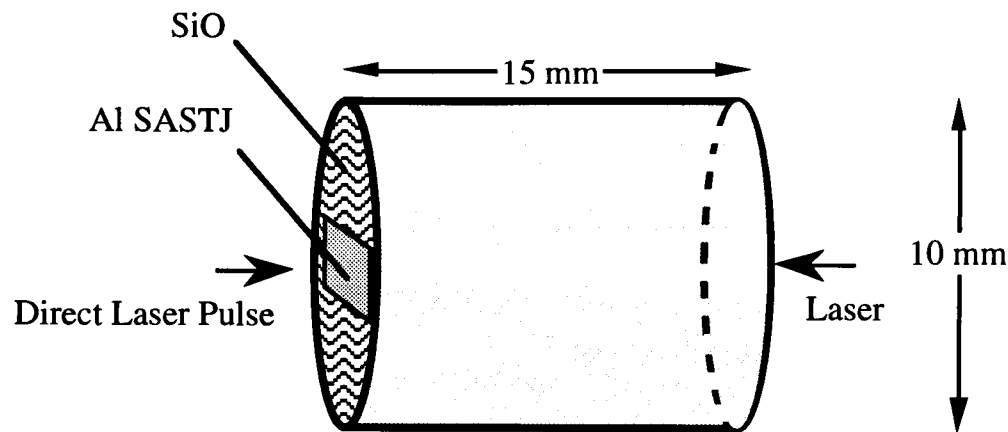


Figure 5.2.1 Diagram of Nb single crystal (15 mm x ø10 mm) with position of laser fibres and SASTJ (see §4.3 for device fabrication details).

Figure 5.2.2 shows the first 12 μs of the voltage signal from an SASTJ in the geometry shown in Fig. 5.2.1 at 500 mK. The voltage signal shape does not directly represent the phonon flux entering the thin films of the junctions (and breaking quasiparticles) due to the integrating effect of the finite quasiparticle recombination time in the Al (in this case, $\tau_r = 1.30 \mu\text{s}$), and the RC effect arising from the electronic characteristics of the SASTJ, leads and head amplifier ($\tau_{RC} = 0.20 \mu\text{s}$). The voltage signal time-base also has a slight delay between the digital oscilloscope trigger and the start of the laser pulse. These effects can be removed satisfactorily by the deconvolution of the signal with an appropriate function representing the response of the SASTJ and electronics to a δ -function injection of quasiparticles directly into the films.⁶⁶ The result of this deconvolution is shown in Fig. 5.2.3. A gaussian time averaging (width 80 ns) was applied to the resulting pulse to improve the signal to noise.

⁶⁶This response can be measured by direct illumination of the SASTJ using a second laser fibre positioned directly above it.

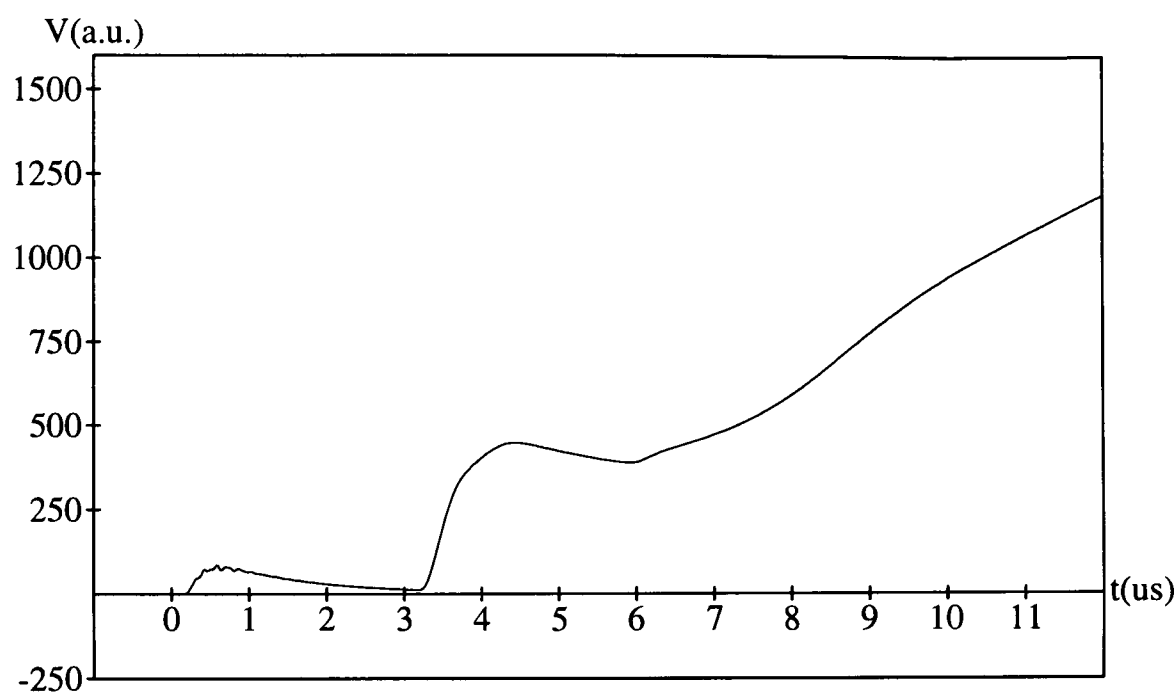


Figure 5.2.2 Voltage signal from a current biased Al SASTJ (shown in Fig. 5.2.1) following a laser pulse on the far face of the Nb crystal.⁶⁷

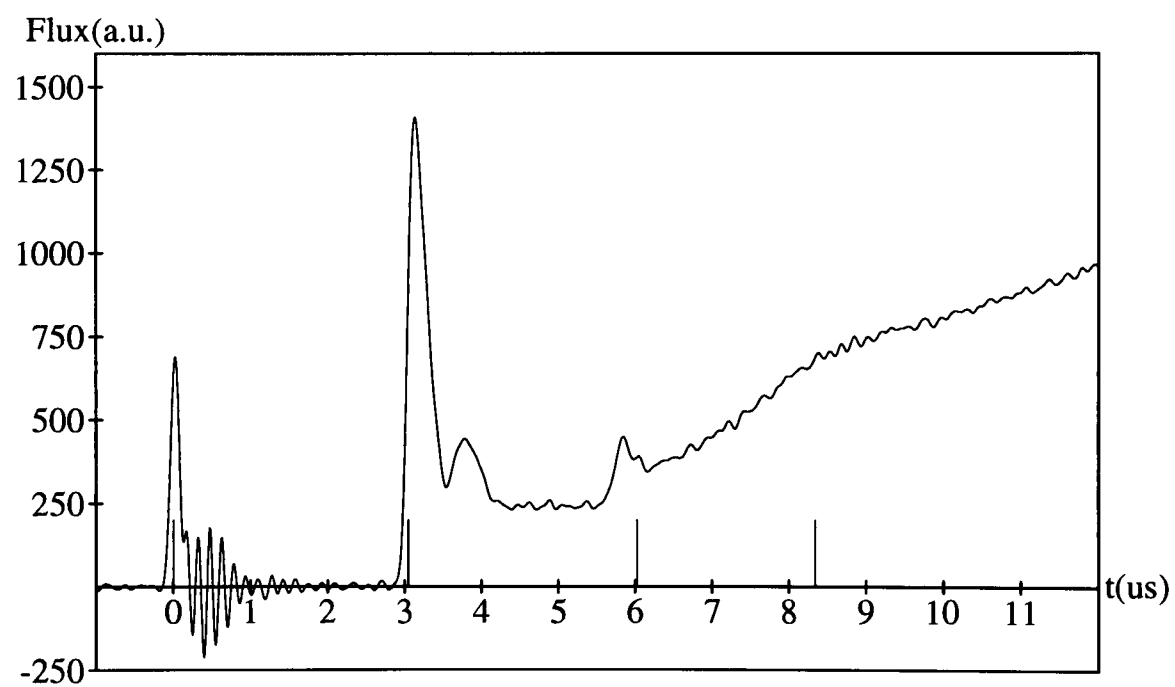


Figure 5.2.3 Signal representing the phonon flux entering a SASTJ, obtained from trace in Fig. 5.2.2 using deconvolution technique outlined in the text. Vertical marks give calculated phonon arrival times, based on elastic constants given in Table 5.2, for the L (5.05 km s^{-1}), FT (2.56 km s^{-1}) and ST (1.85 km s^{-1}) modes.⁶⁸ The spike at $t = 0$ arises due to scattered light from the fibre on the far face falling directly onto the SASTJ.

We will consider the deconvoluted pulse shape in detail. A small signal at $t = 0$ arises due to scattered light from the fibre, aligned with the far face, falling directly on the SASTJ. It's presence as a narrow spike (FWHM $\sim 100 \text{ ns}$) indicates that the

⁶⁷File: "Ballistic Cross Xtal VIIIp47"

⁶⁸File: "Ballistic Cross Xtal VIIIp47"

deconvolution has worked. The following oscillation is electrical pick up from the laser drive mechanism.⁶⁹

The L phonon peak is observed with a leading edge arrival time of $3.06 \pm 0.01 \mu\text{s}$ for the propagation distance of $15.3 \pm 0.1 \text{ mm}$. The error in the distance arises due to the uncertainty in the position of the laser fibre on the far crystal face. This gives a velocity of $5.00 \pm 0.05 \text{ km s}^{-1}$ which is close to that determined by Weber (see Table 5.1) from ultrasonic measurements on Nb. The FWHM of this pulse is $\sim 270 \text{ ns}$. The electronic processing and variation in path length over the area of the SASTJ (100 ns and 75 ns, respectively) contribute the majority of this width. The residual width may arise from the finite lifetime of the mechanism, at the Nb crystal surface, generating the longitudinal phonons.

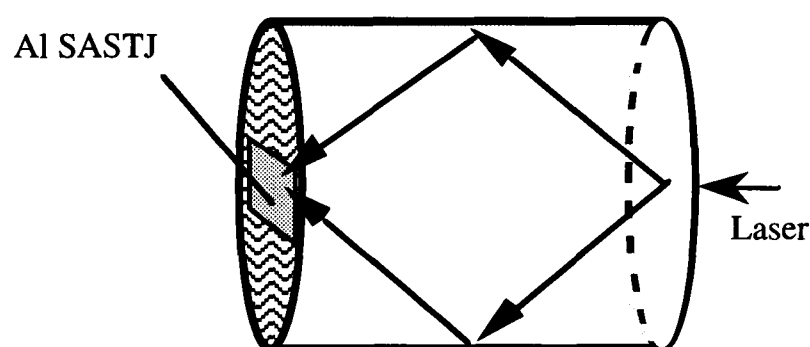


Figure 5.2.4 Paths traced by L phonons causing peak at $3.8 \mu\text{s}$ in the trace shown in Fig. 5.2.3.

The second peak is caused by the reflection of L phonons from the surface of Nb crystal, as depicted in Fig. 5.2.4. It has an arrival time some 18.8% slower than the initial L pulse. This should be compared to an estimate of the additional path length of a reflected signal (for a cylinder diameter of 10 mm) of $\sim 20\%$. The ratio of peak areas for the direct L and reflected L peaks is 1:0.63. Unfortunately, we cannot obtain a precise estimate of the surface reflection coefficient implied by this ratio because of the complexities presented by the cylindrical profile of the crystal combined with an uncertainty in the position of the laser fibre. If as an approximation to the slightly offaxis cylindrical case we assume 100% phonon reflection takes place from four planar surfaces round the surface of the crystal this would predict a ratio of 1:1.4 indicating an actual

⁶⁹It is interesting to note how the deconvolution (Fig. 5.3) emphasises the high frequency pick up when compared to the previous Fig. 5.2

reflection coefficient $\sim 50\%$. A theoretical calculation of the expected reflection coefficient will have to take into account potential mode conversion which would also reduce the L phonon flux [91].

The timing of the reflected pulse also provides evidence for the predicted anisotropy in the propagation speed of L phonons since if our estimate of the additional propagation distance is correct the leading edge appears to travel with a velocity $\sim 5.3 \text{ km s}^{-1}$. We will discuss this in more detail in the next section.

The third peak in Fig. 5.2.3 arises from the [110] FT phonons and corresponds to a velocity of $2.65 \pm 0.03 \text{ km s}^{-1}$. The fourth peak arising from ST phonons appears considerably smoothed out at around $8.5 \mu\text{s}$ ($1.8 \text{ km s}^{-1} \pm 0.1$). The rising background on which it appears is due to reflected FT phonons which arrive before the expected ST peak. As will be discussed in the next section, ST phonons preferentially propagate in the [111] direction (phonon focusing) leading to a reduction in the number observed along the [110] direction.⁷⁰

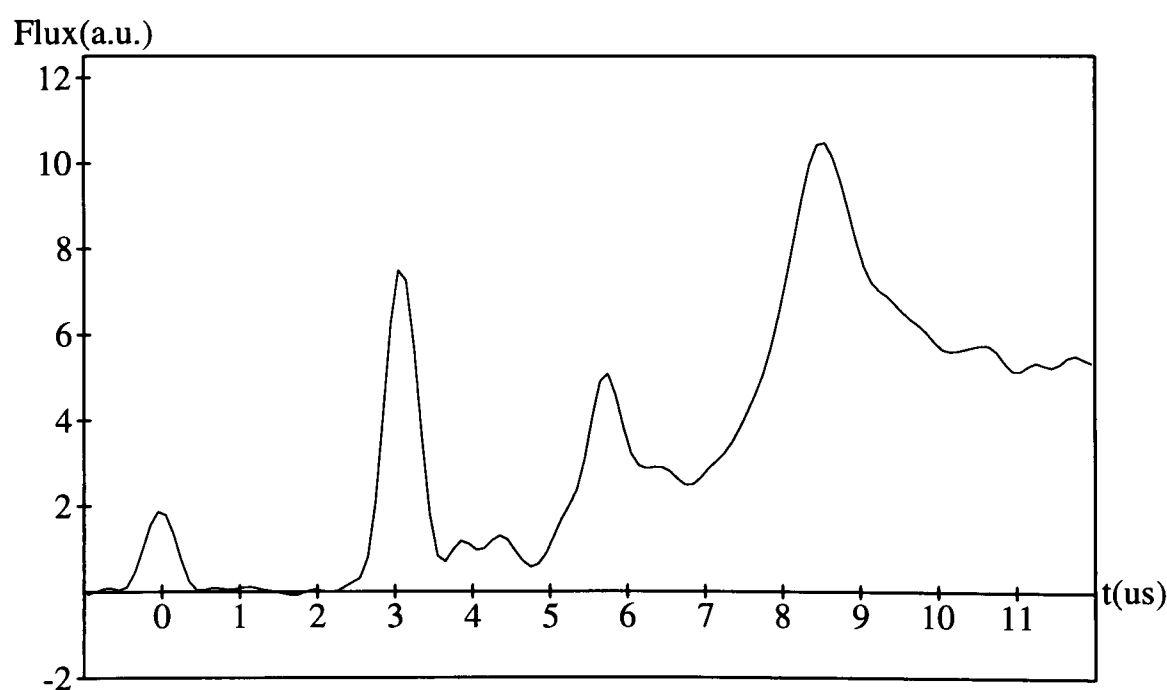


Figure 5.2.5 Signal representing the phonon flux entering a SASTJ obtained using deconvolution technique outlined in the text. A 200 ns gaussian time filter has been applied to improve the signal to noise ratio. The geometry is similar to that shown in Fig. 5.2.1 except that the signal source is from the laser illumination of a Cu-SiO layer on the surface of the Nb crystal. The use of this source of excitation is discussed in more detail at the end of this chapter.⁷¹

⁷⁰Private Communication: B. Cabrera, Stanford, USA.

⁷¹File: "Ballistic Cross Xtal Vp102"

The transverse phonon modes could be observed more readily by using an alternative source of excitation (laser directed at Cu–SiO layer) on the surface of the crystal (see §5.7 concerning the variation of the non–thermal signals in the Nb with excitation source) in a similar crystal geometry to that shown in Fig 5.2.1 (Propagation distance 15.3 ± 0.2 mm). Both the FT and ST peaks can be clearly observed in the deconvoluted pulse shown in Fig. 5.2.5. The ST peak is, once again, smeared out by additional reflected FT phonons. The measured FT propagation speed is 2.67 km s^{-1} which is consistent with the calculated ST velocity at 15° to the [110] direction in the [100] direction. In this case the SASTJ and laser were not arranged symmetrically on the near and far faces.

Mode	Direction	Velocity (km s^{-1})	Reference
L	[111]	4.95	[92]
L	[111]	5.02 ± 0.02	[37](Calc.)
L	[111]	4.943	[86]
T	[111]	2.55	[92]
T	[111]	2.34	[86]
L	[100]	5.41 ± 0.01	[37]
L	[110]	5.10 ± 0.02	[37]
L	[110]	5.00 ± 0.05	(This paper)
FT	[110]	2.65 ± 0.03	(This paper)
ST	[110]	1.8 ± 0.1	(This paper)
ST	[110]	1.865 ± 0.002	[37]

Table 5.2.1 Measurements of the velocities of phonons in Nb at low temperatures.

We will now discuss the phenomena of phonon focusing and also make a calculation of the theoretical phonon velocities along a number of crystallographic axes in Nb based on the published second order elastic constants.

5.2.2. Phonon Focusing

The propagation of elastic waves in crystals can be described in the long wavelength limit by the expression,

$$\rho \frac{\partial^2 u_\alpha}{\partial t^2} = \sum_{\beta\gamma\delta} C_{\alpha\beta,\gamma\delta} \frac{\partial^2 u_\gamma}{\partial r_\delta \partial r_\beta}, \quad [5.1]$$

where ρ is the mass density, $C_{\alpha\beta,\gamma\delta}$ are the second order elastic constants of the crystal and u and r are the local displacement due to the wave and co-ordinates respectively ($\alpha, \beta, \gamma, \delta = 1, 2, 3$). The Nb lattice is a b.c.c. cubic structure. In a cubic system the elastic tensor, $C_{\alpha\beta,\gamma\delta}$, shows a high degree of symmetry and can be best characterised by three independent second order elastic constants c_{11} ($= C_{\alpha\alpha,\alpha\alpha}$), c_{12} ($= C_{\alpha\beta,\beta\alpha} = C_{\alpha\beta,\alpha\beta}$) and c_{44} ($= C_{\alpha\alpha,\beta\beta}$) [10, 93]. The anisotropy⁷² of the elastic constants of the Nb lattice has an interesting effect on the propagation of ballistic phonons in a crystal. The k -vectors and group velocities of the phonons are no longer collinear and the latter vary in magnitude with direction for a given mode. The effect is most pronounced for the FT mode and smallest for the L mode and leads to considerable anisotropy of the phonon flux from a point source within the crystal, concentrating it along certain axes of the crystal. This effect is known as phonon focusing. The elastic constants of Nb have been measured by Alers and also by Weber and are shown in Table 5.2.2.

⁷²The degree of anisotropy of a cubic system can be expressed by the parameter $c^* = c_{11} - c_{12} - 2c_{44}$. c^* will be zero in an isotropic system. The value of c^* in Nb is 53.2×10^{10} dynes cm^{-2} which can be compared with -57.6×10^{10} dynes cm^{-2} in Si.

	c11	c12 (10 ¹⁰ dynes cm ⁻²)	c44	Density (g cm ⁻³)
Alers[38]	254.2	134.0	30.9	
Weber[37]	246 ± 2	134 ± 4	29.4 ± 0.1	8.44 ± 0.01
Values used in calculations	246	134	29.4	8.57 ⁷³

Table 5.2.2 Experimental values of the second order elastic constants of Nb. The last line in the table is the set of parameters used to calculate Figs. 5.2.6–5.2.10.

In Figs. 5.2.6–5.2.8 we have plotted the surfaces, in 3-D, representing the low energy ($\Omega \ll \Omega_D$ ⁷⁴) dispersionless phonon velocity for the three separate phonon modes. The techniques to obtain these surfaces are well documented and will not be discussed further here [10, 93, 95]. In Figs. 5.2.9 and 5.2.10 we have also plotted velocities in the planes through the [100]–[110]–[010] and [100]–[111]–[110] directions. It can be seen that, while the L phonon velocity varies by only 10% in all directions, both the T modes are considerably distorted. The points on the curves of Figs 5.2.9 and 5.2.10 were spaced in *k*-space at 5 degree intervals of θ . Regions where they are closely spaced or the lines double back indicate regions where phonon flux will be concentrated. This can also be clearly seen in the half-octant 3-D projections of Figs. 5.2.6–5.2.8. While the L group velocity surface is close to a sphere, the FT and ST group velocity surfaces are considerably distorted. The fold-back directions, along which phonons will appear to preferentially propagate, are the [111] directions for ST phonons, and the [111] and (more significantly) the [110] directions for the FT mode.

In high purity Nb single crystals low energy phonons ($\Omega \approx 400 \mu\text{eV}$) propagate ballistically with m.f.p.'s of many cm s. The group velocity surfaces of the phonons are extremely complex and warrant further study. Given the limited flexibility of our current laser system in our cryostat we have been unable to map out these focusing patterns. However, the anisotropy of the phonon velocities were confirmed for a number of fixed

⁷³Density at 300 K from [94]. Thermal contraction at low temperatures would increase this value ~1%.
⁷⁴ $\Omega_D (= k_B \Theta_D)$ is the Debye energy which is around 23.7 meV in Nb averaged over all phonon modes.

laser positions. The importance of these focusing patterns will have a number of ramifications for the detection of the position of particle interactions within the bulk of Nb crystals and will be discussed in §7.

Table 5.2.3 contains the expected phonon velocities as calculated from the elastic constants given in the last line of Table 5.2.2. As can be seen there is good agreement with those experimental values obtained for propagation in the [110] direction.

Mode	Direction	Phonon Velocity ⁷⁵ (km s ⁻¹)	ρV^2
L	[100]	5.36	c_{11}
T	[100]	1.85	c_{44}
L	[110]	5.06 (5.00 ± 0.05)	$(c_{11}+c_{12}+2c_{44})/2$
T1[001] ⁷⁶	[110]	1.85 (1.8 ± 0.1)	c_{44}
T2[110̄]	[110]	2.56 (2.65 ± 0.03)	$c_{11}-c_{12}$
L	[111]	4.96	$(c_{11}+2c_{12}+4c_{44})/3$
T	[111]	2.35	$(c_{11}-c_{12}+c_{44})/3$

Table 5.2.3 Calculated phonon velocities using parameters given in last line of Table 5.2.2. These values compare well with those we observed experimentally (shown in brackets).

⁷⁵Experimental values determined in this work shown in brackets.
⁷⁶Direction of transverse displacement

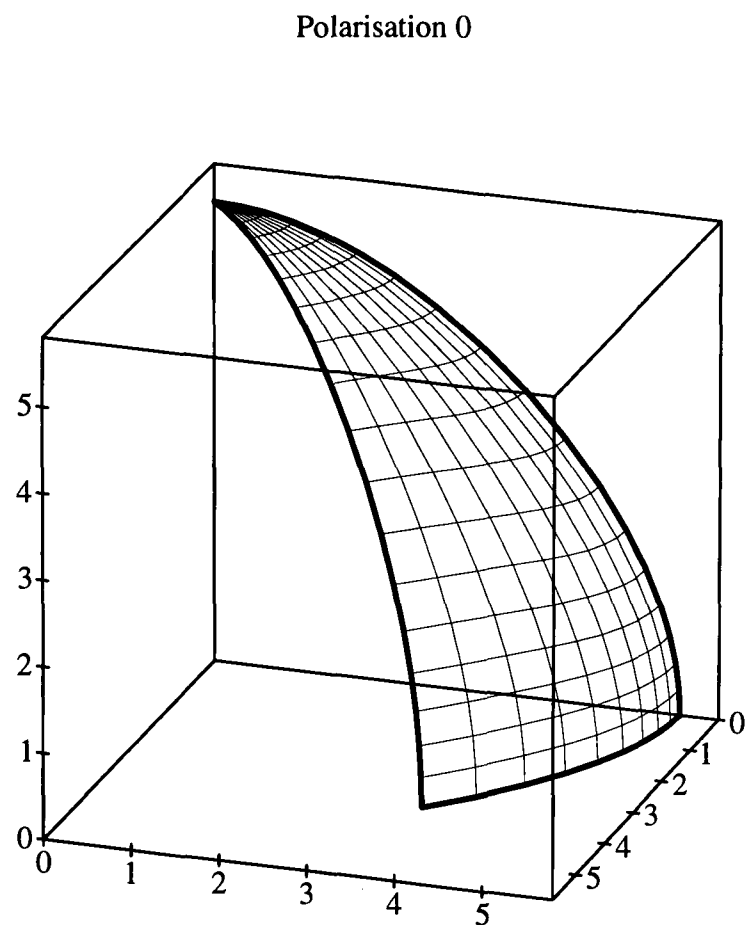


Figure 5.2.6 Longitudinal phonon group velocity (km s^{-1}) in Cartesian co-ordinate system. Only half of one octant mapped from k -space is plotted for simplicity. The high degree of symmetry means that the full surface can be created by the reflection of this surface into the other sectors. The lines are spaced in 5 degree intervals of θ and ϕ in k -space. Areas where they are more closely spaced compared with the normal isotropic pattern (or where they double back) indicate directions of increased phonon focusing.⁷⁷

⁷⁷File: "Focus/Group Vel in Nb 19.12.92"

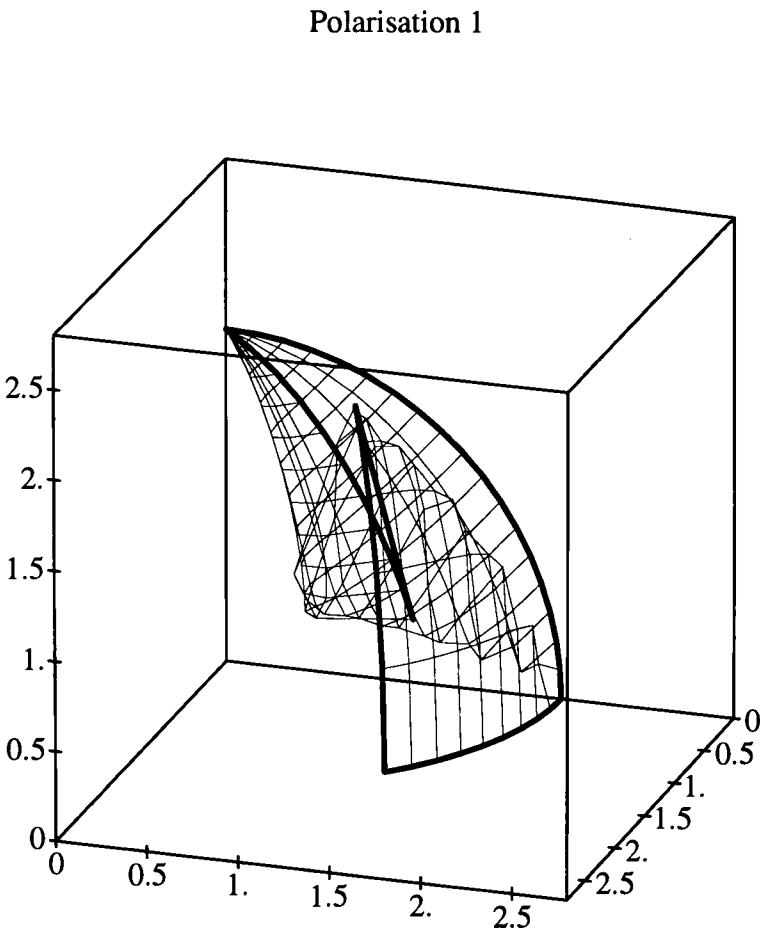


Figure 5.2.7 Slow transverse phonon group velocity surface (see caption of Fig. 5.2.6).

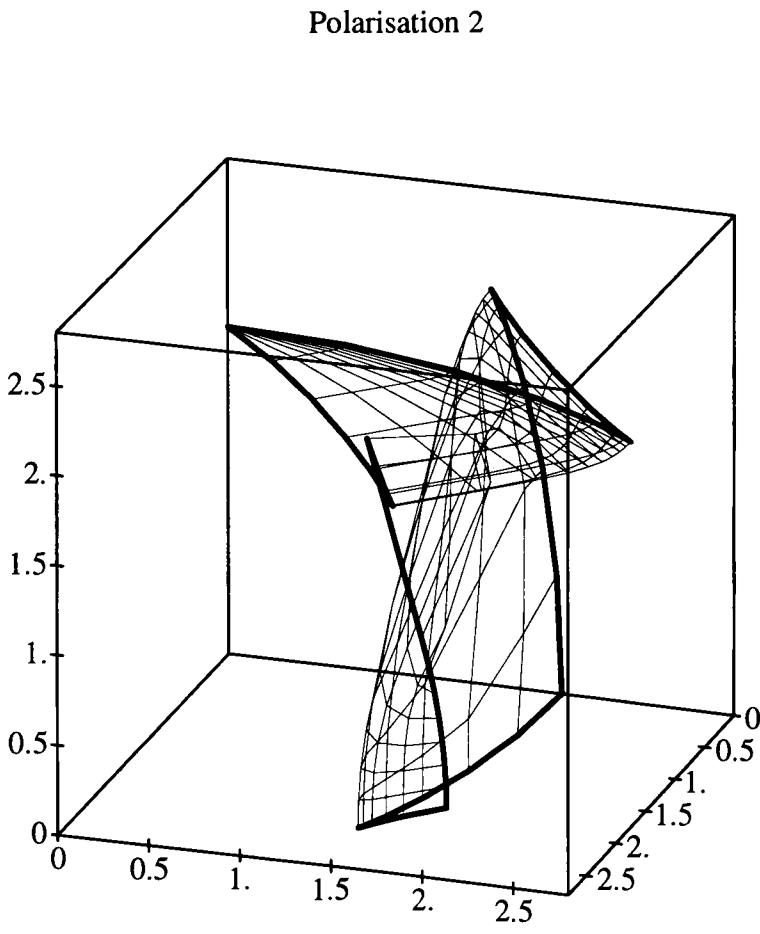


Figure 5.2.8 Fast transverse phonon group velocity surface (see caption of Fig. 5.2.6).

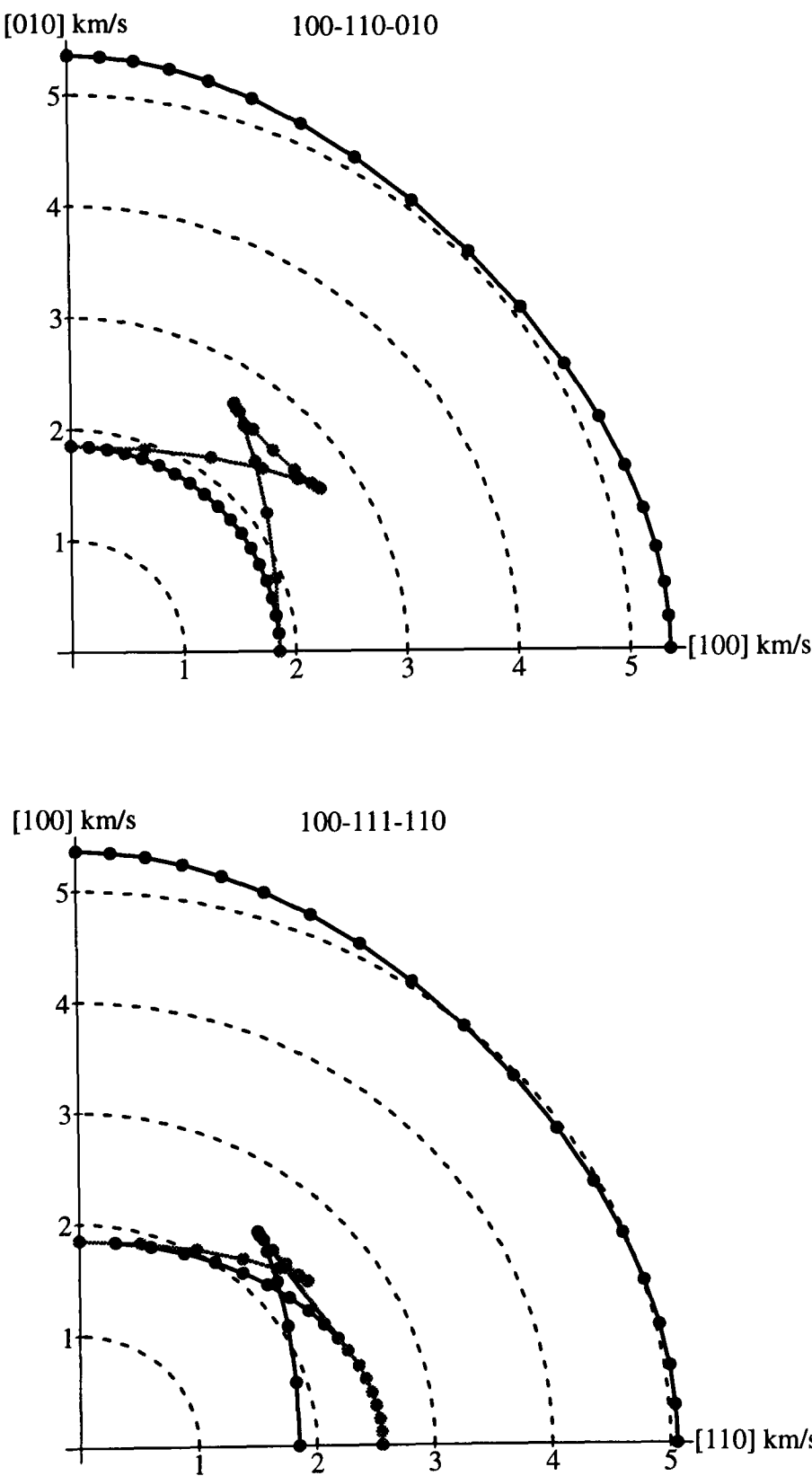


Figure 5.2.9 and 5.2.10 Phonon group velocities in the planes containing the directions indicated. The L phonon velocity varies by less than 10% in all directions leading to minimal focusing effects. The FT (grey line) and ST (black inner line) modes show considerable anisotropy. The major focusing directions are along the [110] and [111] directions. The points are spaced at 5 degree intervals in θ in k -space.⁷⁸

⁷⁸File: "Focus/Group Vel in Nb 19.12.92"

5.3. TEMPERATURE DEPENDENCE OF PHONON MEAN FREE PATH DUE TO QUASIPARTICLE SCATTERING

Early experiments on single crystal Nb were conducted on low purity samples ($RRR \sim 10$) of thicknesses 1–2.5 mm. STJs were fabricated on the surface in Nb–SiO–Al–Ox–Pb, Nb–SiO–Al–Ox–Al and Nb–Ox–In geometries as illustrated in Fig. 5.3.1 and 4.3.1. The threshold for phonon detection in these junctions is set by the film with the lowest superconducting gap ($\Omega \geq 2\Delta$).

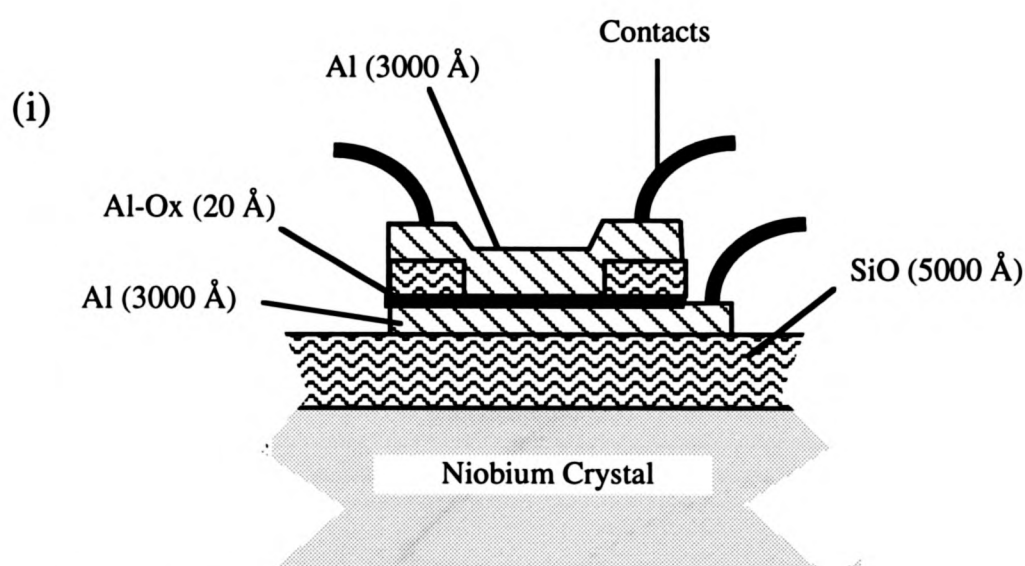


Figure 5.3.1 Schematic of Nb–SiO–Al–Ox–Al STJ on the surface of Nb crystals

The voltage signal shapes detected, in the first 50 μs , following laser excitation of the far face of the crystal, were consistent with the diffusion of phonons across the crystal. At temperatures below 1 K the shape of diffusion signal was independent of temperature. In the low purity Nb crystals the phonon m.f.p. saturates due to scattering from impurities within the crystal. Figure 5.3.2 shows a typical phonon pulse shape observed in a Nb–SiO–Al–Ox–Pb STJ at 500 mK. The shape of the pulses could be fitted using the diffusion model ⁷⁹ outlined in §3.5 with a diffusion coefficient of $0.15 \text{ m}^2\text{s}^{-1}$ and a reflection coefficient ~ 1 (although the signal shape, in this case, is relatively insensitive to the reasonable values of the latter parameter). For a thin crystal (propagation distance $\ll$ diameter of the crystal) the solutions of this model (0–40 μs) are identical to those of the simple 3–D diffusion equation (Eq. 3.5.1),

⁷⁹After allowing for the response of the STJ, as detailed in §3.4.3.

$$\psi(\vec{r}, t) = \frac{1}{(4\pi Dt)^{\frac{3}{2}}} e^{-\frac{|\vec{r}|^2}{4Dt}}, \quad [5.3.1]$$

where D is the diffusion coefficient, t is the time and $\vec{r}$ is the position of the point relative to the source (assumed a δ -function in space and time). The peak maximum of this function is at a time, t_{max} ,

$$t_{max} = \frac{r_{prop}^2}{6D}, \quad [5.3.2]$$

for a propagation distance, r_{prop} .

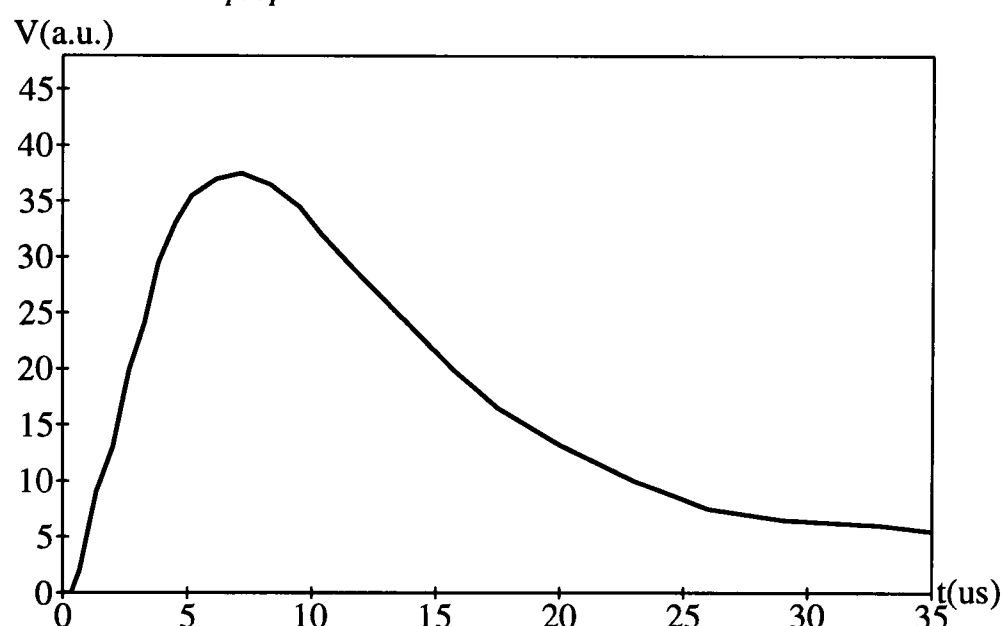


Figure 5.3.2 A typical voltage pulse shape detected in a Al-Ox-Pb STJ on the insulated surface of a low purity Nb crystal following laser excitation of the far face of the crystal. The signal is due to the diffusion of phonons over 2.4 mm across the crystal.

The diffusion coefficients of the pulses can be obtained directly from Eq. 5.3.2 after adjusting for the STJ response function. A plot of the temperature dependence of the diffusion coefficient observed in a number of Nb samples is shown in Fig. 5.3.3. At temperatures above 1.5 K the phonon diffusion coefficient decreases rapidly with increasing temperature. This is due to the scattering of phonons from thermally generated quasiparticles within the Nb. The line is calculated using the relation

$$D = \frac{1}{3} v_{\phi} \Lambda_{\phi}. \quad [5.3.3]$$

where v_{ϕ} is the phonon velocity. Λ_{ϕ} is the phonon m.f.p. which is given by

$$\Lambda_{\phi}^{-1} = \Lambda_{\phi qp}^{-1} + \Lambda_{imp}^{-1}, \quad [5.3.4]$$

where Λ_{imp} is the impurity scattering length and $\Lambda_{\phi qp}$ is the path length for scattering from thermally generated quasiparticles. The phonon–quasiparticle scattering rate⁸⁰ is given by Eq. 3.3.25 which for $\Omega \ll k_B T$ and Δ_{Nb} , simplifies to

$$\tau_{\phi qp}^{-1}(\Omega) = (\tau_0^{ph})^{-1} \frac{2\Omega\pi}{\Delta(0)} f(\Delta(T)). \quad [3.15]$$

τ_0^{ph} is the characteristic scattering time of phonons from electrons in the normal state. The presence of the Fermi function reflects the reduction of the number of unbound electrons (due to the formation of Cooper Pairs in the superconductor) when we move from the normal to the superconducting state.

If we take the value of $\tau_0^{ph} = 4.17$ ps for Nb from Kaplan[36] and use the transverse phonon velocity ($v_\phi = 1.85$ km s⁻¹) such that

$$\Lambda_{\phi qp}(\Omega, T) = v_\phi \tau_{\phi qp}(\Omega, T), \quad [5.3.6]$$

we can evaluate Eq. 3.3.25⁸¹. We assume an average phonon energy near the threshold of the STJs ($\Omega \sim 400$ μ eV) and find that an impurity limited phonon m.f.p., Λ_{imp} , ~ 100 μ m is best able to fit the low temperature data. This is consistent with the previous measurements of Pannetier et al[92].

⁸⁰For phonons below the $2\Delta_{Nb}$ threshold.

⁸¹Bobetic[51] also gives an expression for the ratio of normal state to superconducting mean free paths which is equivalent to our analysis

$$\begin{aligned} \frac{\Lambda_N}{\Lambda_S} &= \frac{1}{\Omega} \int_{\Delta}^{\infty} \frac{dE (f(E) - f(E + \Omega)) (E(E + \Omega) - \Delta^2)}{(E^2 - \Delta^2)^{\frac{1}{2}} ((E + \Omega)^2 - \Delta^2)^{\frac{1}{2}}} \\ &= \frac{2}{\Omega} \int_{-\infty}^{\infty} dE \left(1 - \frac{\Delta}{EE'}\right) (f(E) - f(E')) \rho(E) \rho(E') \\ &\approx 2f(\Delta(T)) \text{ for } \Omega \ll \Delta \end{aligned}$$

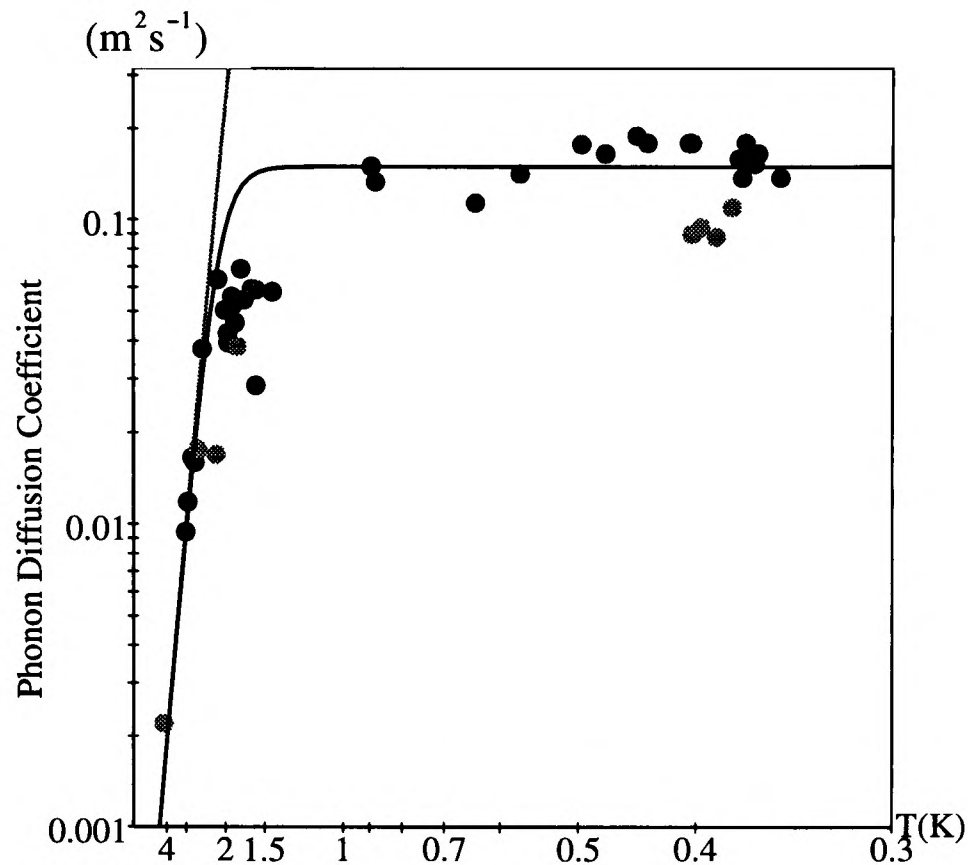


Figure 5.3.3 Diffusion coefficient of phonons (m^2s^{-1}) as a function of temperature measured in a number of low purity Nb crystals over propagation distances ranging from 1–3 mm. On the horizontal axis the temperature T is scaled as T^{-1} .⁸²

Using the greater sensitivity of an Al SASTJ we were able to confirm that small additional structure was present at the leading edge of the diffusion pulses which was consistent with ballistic phonon transport. This feature was $\ll 1\%$ of the phonon signal contained in the diffusion pulse.

5.4. QUASIPARTICLE PROPAGATION

5.4.1 Introduction

In §5.2 and 5.3 we discussed observations of the ballistic and diffuse propagation of phonons in the high and low purity Nb crystals, respectively. In §5.5 we will detail further experiments performed on the high purity crystals that look at the behaviour of the non-equilibrium signal at temperatures below 2 K. For reasons that will become apparent we have chosen to discuss the propagation of quasiparticles separately in this section.

⁸²File: "Plot Diff Coeff vs 1/T 6.1.93"

In principle, a non-equilibrium excess of quasiparticles in a bulk superconductor can be detected in two ways. Firstly, by using a STJ formed with the crystal as one of the electrodes, shown schematically in Fig. 5.3.2(ii). Quasiparticles within about a coherence length of the oxide barrier on the surface of the crystal can tunnel through the S–I–S junction giving an additional tunneling current. Secondly, the quasiparticles can be detected indirectly by observing their recombination phonons (see §3.3.2 and 3.3.3) in a STJ which is insulated from the crystal surface. The efficiency of this second process is reduced as the temperature is lowered due to the $e^{-\Delta/k_B T}$ dependence in the quasiparticle recombination rate in the crystal.

The results of Pannetier [28] demonstrated quasiparticle detection using the second technique. He observed the extinction of the mixed-mode of propagation (see Fig.3.3.3) as the temperature was lowered. No pure quasiparticle mode was observed.⁸³

Narayanamurti *et al.* [27] and Goldie [31] were able to detect the quasiparticle pulse in Pb and In single crystals directly and found that it was eliminated by a thin layer of SiO between the crystal and detector. In both these cases the crystal surfaces were cleaned immediately prior to junction fabrication.

5.4.2. Direct Observation of Quasiparticle Propagation

We were able to observe the non-equilibrium quasiparticle signals in Nb using Nb–Ox–Al and Nb–Ox–Pb junctions made directly on the crystals.

In the low purity Nb crystals (RRR~10) the phonon diffusion pulse (shown in Fig. 5.3.2) was followed by a second diffusion pulse with a characteristic diffusion coefficient some 20 times lower ⁸⁴. Figure 5.4.1 shows the voltage signal from an Nb–Ox–Al

⁸³In order to fabricate a Sn–Ox–Sn STJ with the Sn crystal forming the lower electrode, Pannetier deposited a thin Sn film on the surface of the crystal prior to oxidation. It is apparent that the quasiparticles were not being transmitted from the bulk crystal into this thin film and therefore were not able to tunnel through the oxide barrier into the second Sn probe film. The pulse shapes from a second device with a Sn–Ox–Sn STJ separated from the crystal by 2000 Å of SiO were identical to those seen for the first type of junction.

⁸⁴The diffusion coefficient was $\sim 6 \times 10^{-3} \text{ m}^2\text{s}^{-1}$ which correspond to a m.f.p. of 1000 Å. A four fold improvement of the quasiparticle diffusion coefficient in the low purity crystals was achieved by a short period of vacuum out-gassing at 1000 °C.

junction at 360 mK. The Al film has a threshold for phonon detection⁸⁵ of $\Omega \geq 340 \mu\text{eV}$ and so the STJ is sensitive to both the phonon and quasiparticle signals. In similar experiments performed with Nb–Ox–Pb junctions the magnitude of the phonon pulse was significantly reduced [96] (relative to the quasiparticle signal) due to the much higher threshold for phonon detection $\Omega \geq 2700 \mu\text{eV}$ in these devices.

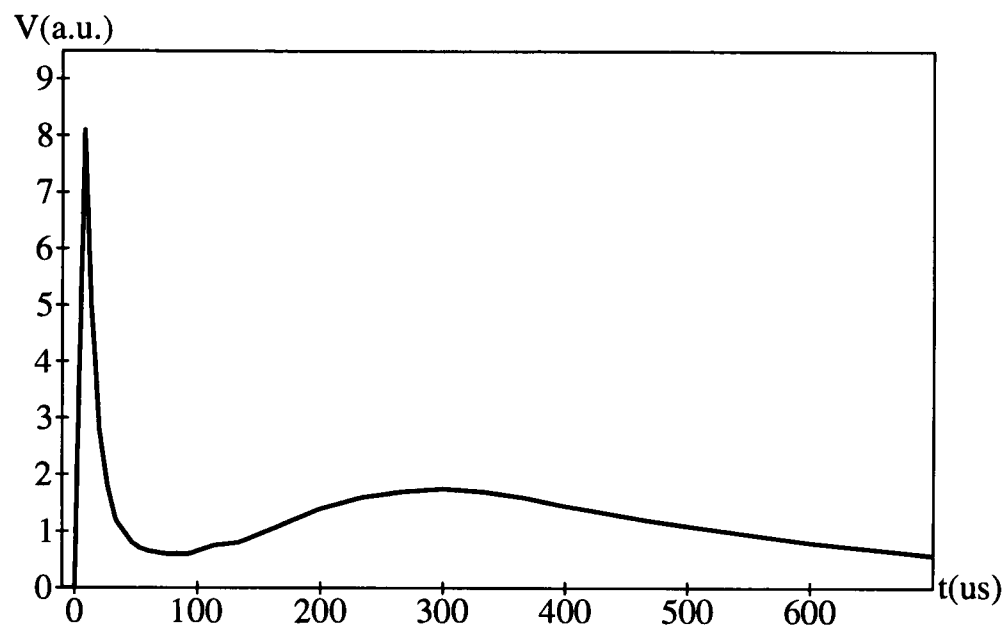


Figure 5.4.1 Voltage signal from a Al–Ox–Nb STJ following laser illumination of the far face of a low purity Nb crystal (length 2.5 mm, diameter 10 mm). The leading peak is due to phonons and the following signal due to quasiparticles. The ac coupling time of the amplifier rolls the signal off at long time-scales.

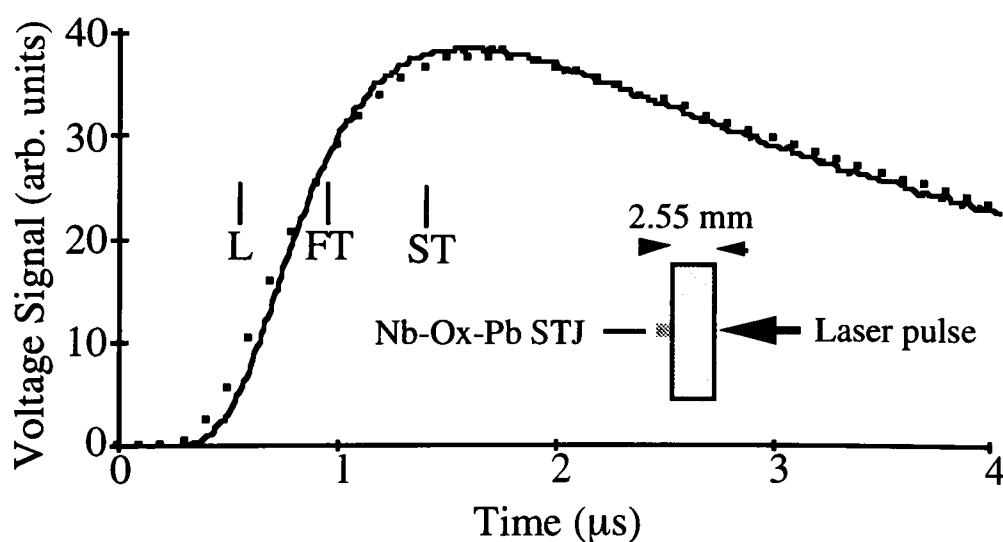


Figure 5.4.2 Quasiparticle signal, detected in Nb_{crystal}–Ox–Pb junction at 400 mK, propagating 2.55 mm in [110] direction. Dotted line is fit for diffusion coefficient $0.7 \text{ m}^2\text{s}^{-1}$. For comparison the timings for the three ballistic phonon propagation modes (L, FT, ST), over the same distance, are indicated.

⁸⁵Phonons of energy $\geq 2\Delta_{\text{Nb}}$ are strongly attenuated in the Nb crystals due to pair breaking (see §3.3.6).

The experiments outlined in the last paragraph were repeated on the high purity Nb crystals ($RRR \sim 10^4$). Figure 5.4.2 shows the signal detected at 400 mK by a Nb–Ox–Pb junction. The pulse shape is consistent with a three orders of magnitude improvement in the quasiparticle m.f.p. A fit with our diffusion model (see §3.5), after adjusting for the response of the junction, gives a diffusion coefficient of $0.7 \text{ m}^2\text{s}^{-1}$. The leading edge of the signal was found to be just in advance of that expected for L phonon propagation. The expected phonon arrival times are marked in Fig. 5.4.2)⁸⁶. The signal from a Nb–Ox–Al junction on the same device showed additional structure in the pulse that was consistent with the detection of these ballistic phonon modes.

In the range of temperatures (0.36–1.2 K) that we were able to detect the quasiparticle signal (above the electronic noise) no variation in the diffusion coefficient was observed⁸⁷. This was surprising since the velocity of quasiparticle propagation is expected to be temperature dependent, due to the curvature of the energy–momentum dispersion relationship around the Fermi momentum, according to the relationship [27]

$$v = \left(\frac{2k_B T}{\pi \Delta} \right)^{\frac{1}{2}} v_F, \quad [5.4.1]$$

where v_F is the Fermi velocity. However, this relationship will not hold if the excess quasiparticle number dominates the thermal population in the crystal. Calculations of the laser energy indicate that the experiment was in this regime leading to a much weaker temperature dependence in the velocity. In addition, the rather complex nature of the Nb Fermi surface may also lead to more complex behaviour of the quasiparticle velocities.

5.4.3 Ambiguities in the Quasiparticle Decay Times

In our initial investigations [96] of phonon and quasiparticle propagation in Nb we attributed the observation of a long lived tail signals (5–100 ms) to quasiparticles. This

⁸⁶In the investigations of single crystal Sn, Pb and In (listed in Table 3.1.1) the times of the leading edges of quasiparticle signals over $\sim 2.5 \text{ mm}$ were less than 100 ns. The diffusion coefficient of the quasiparticles in these materials were typically one to two orders of magnitude better than that determined for our high purity Nb samples. This is in part due to the Fermi velocity of Nb being 10 times lower than that in the other materials.

⁸⁷The diffusion coefficient is given by $D = \frac{1}{3} v \lambda$ where v is the propagation velocity and λ is the m.f.p.

tail signal is present both in junctions directly on the crystal and geometries where the S–I–S junctions were insulated from the crystal. The latter type of STJ would be sensitive to the phonons emitted by quasiparticle recombination. The long decay times were independent of temperature in the range 0.36–1.2 K ⁸⁸. This would be expected since the effective quasiparticle lifetime (see §3.3.3) due to thermal recombination would be in excess of 1 s below 1.2 K. The recombination time could be limited by other mechanisms, such as recombination in damaged or trapped flux regions at crystal surface ⁸⁹.

Subsequent analysis of the temperature dependencies of the tail signal heights were difficult to interpret in terms of the tail being due to quasiparticle recombination. A possible model is given in Appendix C, however, another model was proposed in terms of the phonon heat capacity of the crystals and their effective heat sinking (see §5.6). The simplicity of this model and the absence of free parameters seem to indicate that it may be closer to the truth. If this is the case then there are two possible interpretations of the data to date. Firstly, that the quasiparticles are recombining at time scales shorter than the heat sinking time but we are unable to explicitly separate this time constant from the complex behaviour of the non–equilibrium phonon signal (see §5.5.4). Secondly, that the quasiparticles are recombining so slowly that within the current limits of signal to noise we are unable to identify a characteristic decay time beyond that of the thermal time constant ⁹⁰.

5.4.4. Quasiparticle Trapping

Booth [97] has proposed an additional scheme for the detection of quasiparticles. Excess quasiparticles created in a superconductor (in this case, a Nb single crystal) may be trapped into an adjacent volume of a smaller gap superconductor. Figure 5.4.3 shows a

⁸⁸The signals became obscured by electronic noise above this range.

⁸⁹A saturation of the quasiparticle lifetimes at low temperatures was also observed in single crystal In

($\tau_{eff} = 37 \mu s$) [31]. No definitive explanation has yet been provided for this effect.

⁹⁰It should be remembered that at short time–scales we have identified quasiparticle propagation using junctions directly on the crystal. The difficulty lies in establishing the quasi–particle decay time. Unfortunately, this decay time is closely linked to the efficiency with which we can detect them using STJs not in direct contact with the crystal.

schematic of this process. The quasiparticles in the large gap material S_1 have energies near to the energy gap Δ_1 . If these quasiparticles move into S_2 where the energy gap is Δ_2 is smaller, they can scatter by emitting a phonon (see §3.3.4) to lower energies near the energy gap Δ_2 . The quasiparticles now have insufficient energy to return to S_1 and are trapped. The amplitude of the excess tunneling current in an STJ made directly on the trap will now be determined by the volume of S_2 . Booth shows that, by a suitable choice of superconductors and volume ratios, significant improvements in the detected signal is possible ⁹¹.

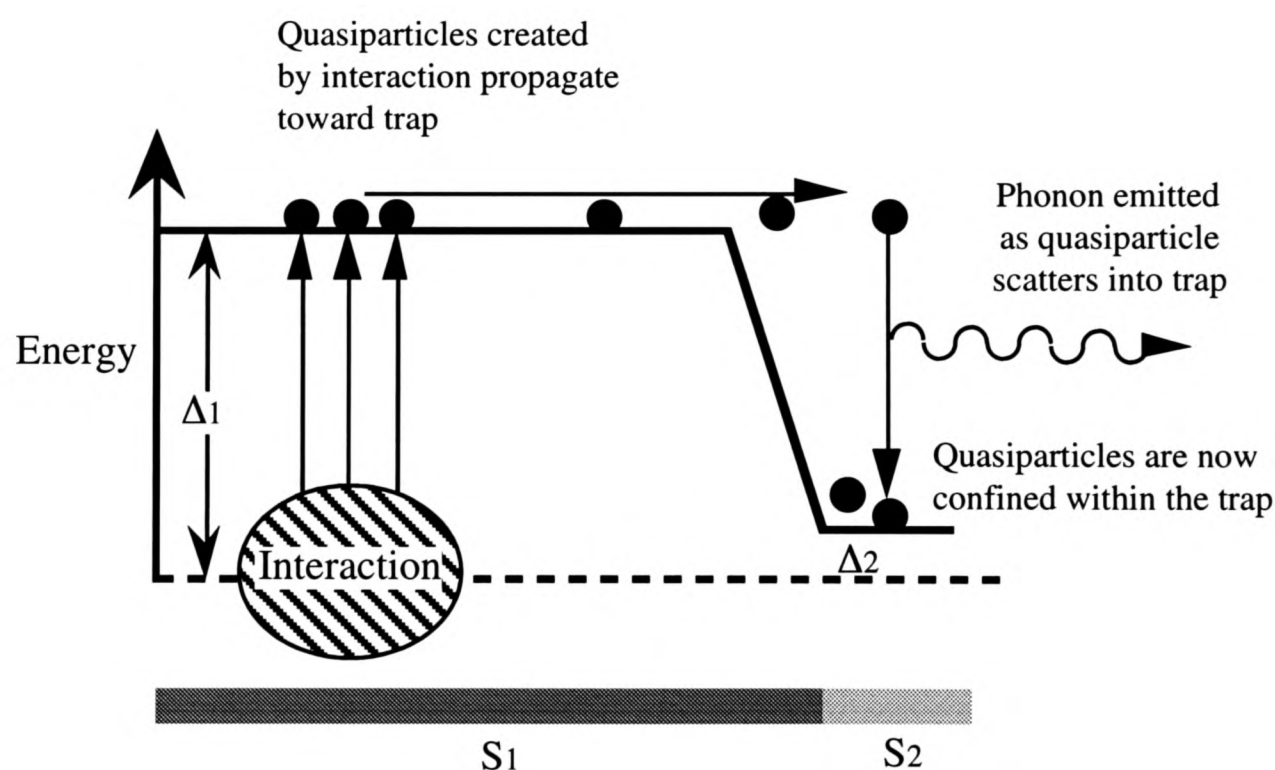


Figure 5.4.3 Quasiparticle trapping [97].

The trapping of the quasiparticles can also be observed by detecting the phonons emitted as the quasiparticles relax into the lower energy gap material [98, 99]. A S–I–S junction (energy gap Δ) above the trap but insulated from the crystal will be sensitive to these phonons as long as $2\Delta \leq \Delta_2 - \Delta_1$.

Our attempts to demonstrate quasiparticle trapping from Nb single crystals were frustrated by low transmission coefficients at the interface between the crystal and the trap. The traps were formed by an Al thin film (60–2000 Å) deposited on one face of the

⁹¹The quasiparticle tunneling rate (and current) in an STJ is inversely proportional to the volume of the superconductor from which they are tunneling. See Eq. 3.4.3 and 3.4.4.

crystal. The quality of the vacuum in our film evaporation system was found to be too poor to maintain a clean Nb surface during the period between ion milling and subsequent Al deposition (see §6.3). We have been able to significantly improve the quality of the interface by moving fabrication to a CVC sputtering system with a much higher quality vacuum. At present we are investigating quasiparticle trapping from these devices.

It should be mentioned that in his original paper Booth gave the time taken to trap the excess quasiparticles from S_1 into S_2 as

$$\tau_{trap} = \frac{V_1}{V_2} \tau_s$$

where V_1 , V_2 are the volumes of the two superconductors and τ_s is the time for the scattering process to lower energy in S_2 (which is typically ~ns). This relationship holds only if the characteristic propagation time of the quasiparticles around V_1 is small compared to τ_{trap} . In Nb the diffusion coefficient is such that, for 2–12g crystals, this condition is not met. The transmission probability at the crystal–trap interface, combined with the diffusion coefficient of the quasiparticles, will limit the trapping time.

5.4.5. Conclusion

The results to date for the observation of quasiparticle propagation in Nb are limited. We have identified the significant improvement in m.f.p. that was available by increasing the purity of the Nb. However, our high purity crystals are probably as pure as is currently possible. In these crystals the propagation times for the quasiparticles and phonons are very similar over distances from 2–10 mm. This makes identification of the relative components of the signals from an interaction difficult.

The results reported in §5.5 and 5.6 for Al–Ox–Al and Sn–Ox–Sn junctions on the insulated surface of the Nb will be discussed, in the most part, in terms of the dynamics of the non–equilibrium phonon signals. We believe that part of the signal may also be due to quasiparticles which recombine at the surface of the crystal, however, we have yet to identify this contribution unambiguously. We hope to perform experiments using

quasiparticle trapping (and phonon spectroscopy) in the near future that will identify their more contribution clearly.

5.5. PHONON PROPAGATION AT LOW TEMPERATURES ($T \leq 2$ K) IN HIGH PURITY NIOBIUM CRYSTALS

5.5.1. Introduction

In this section we will discuss the temperature dependence of phonon signals observed in Al-Ox-Al STJs and SASTJs fabricated on the surface of high purity Nb single crystals. The junctions were insulated from the crystal surface by a thin layer of SiO. We will also present results from a Sn-Ox-Sn STJ fabricated on the anodised surface of a Nb crystal. Excitations were again generated on the far face of a Nb crystal using a laser. The signal propagation distances were 5–10 mm for the STJ work and 15 mm for the Al SASTJ. The electronics and post-processing digital filters were arranged to remove the effects of AC coupling in the voltage pulses from the STJs, while maintaining a high band-width (50 MHz). Thus we were able to analyse the ballistic phonon features on the leading edge (3–10 μ s) and the subsequent evolution of the signal out to ~ 2 ms.

We were able to confirm that in the temperature range 0.3–1.0 K that there is no variation in the attenuation of L or T ballistic phonons. This is in agreement with a calculation of the expected scattering rate from thermally generated quasiparticles (see §5.3) which would become comparable with the propagation distance (10 mm) at 1.5 K for $\Omega = 400$ μ eV. Separate measurement of the variation in the flux of L ballistic phonons generated by a number of fibres along the edge of a crystal suggested that the m.f.p. due to impurity scattering was at least comparable to the dimensions of the crystals. Uncertainties in the yield of ballistic phonons from each of the fibres prevented a more accurate determination of the low temperature m.f.p.

At time-scales ~ 1 ms it was shown that the “non-equilibrium” STJ voltage signals (for constant current bias) corresponded to those expected for a thermal shift in their IV

characteristics. In each case the temperature rise of the crystal was calculated using the Debye heat capacity of Nb and an estimate of the total energy absorbed by the crystals from the fibre pulse⁹². This is the first time that a quantitative analysis has been made of the behaviour of STJs over ms time-scales following a non-equilibrium excitation of a crystal on which they are fabricated. Indeed, once we have a sound understanding of the way in which an STJ responds to changes in the substrate phonon spectrum we can draw conclusions about the behaviour of the phonons within the crystals themselves.

Interpretation of the shape of the STJ voltage signals in the region after the ballistic signal, $\sim 10 \mu\text{s}$, out to 0.5 ms, requires a detailed understanding of the mechanisms responsible for up and down-conversion of the non-equilibrium phonons, within the Nb crystals.

The analysis of the phonon pulse shapes is further complicated by the variation of phonon m.f.p. in Nb with their energy, Ω . Phonons above a threshold of $1200 \mu\text{eV}$ (see §5.5.3) were observed to propagate in the Nb with a diffusion coefficient of $0.5 \text{ m}^2\text{s}^{-1}$ and with an apparent lifetime of $20 \mu\text{s}$. The expected energy dependence of phonon m.f.p. (due to impurity scattering) and lifetime (due to anharmonic decay) are Ω^{-4} and Ω^{-5} (see §3.3.9) [53, 100] and so this is consistent with the observation of ballistic propagation of phonons (over distances $\sim 10 \text{ mm}$) at the Al film threshold ($2\Delta_{\text{Al}} \sim 400 \mu\text{eV}$).

In the following sections we will discuss the observed temperature dependence of non-equilibrium phonon signals in Nb crystals. This will also involve the experimental verification of the theories, developed in §3, concerning STJ sensitivity to non-equilibrium and thermal equilibrium phonon distributions. We will also demonstrate the significant enhancement in sensitivity to phonon signals available by using SASTJs. In §7 we describe the use of these devices to detect the non-equilibrium phonon signals created by the interaction of alpha and gamma particles.

⁹²This estimate of the energy absorbed by the Nb crystals ($\sim 10 \text{ GeV}$) following the laser excitation was in agreement with the SASTJ signal sizes for the interaction of an alpha particle of energy 6 MeV .

5.5.2. Detection using Aluminium Superconducting Tunnel Junctions

An Al–Ox–Al STJ was fabricated on the insulated surface of a 10 mm long, 10 mm diameter Nb crystal and cooled in the ^3He cryostat in the geometry shown in Fig. 5.5.1. The junction was formed at the overlap of two 2500 Å Al films giving a tunneling area of $300\text{ }\mu\text{m} \times 180\text{ }\mu\text{m}$ with a normal state resistance of $1.6 \times 10^{-5}\text{ }\Omega\text{cm}^2$ (30 m Ω). A high conductance junction was used in order to reduce electronic RC effects.⁹³

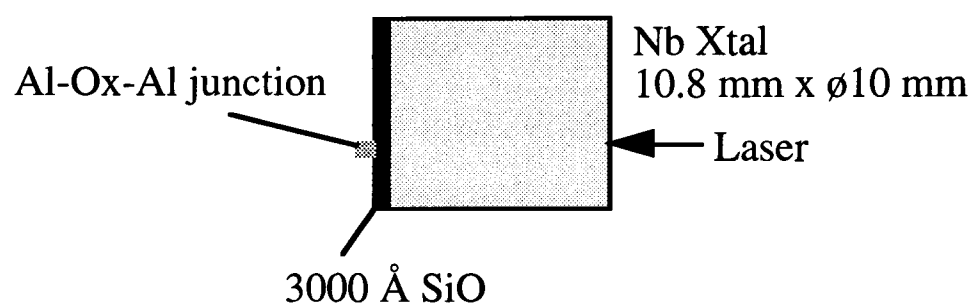


Figure 5.5.1 Geometry of Al–Ox–Al junction on Nb crystal.

Due to variation in the evaporator base pressure during the STJ fabrication the upper and lower Al films had different energy gaps and T_c 's, 210 μeV , 1.67 K and 160 μeV , 1.22 K. The measurement of the energy gaps from the IV characteristic slightly underestimates the gap due to junction self-heating.

The Al STJ will have a detection threshold for phonons ($\Omega \gtrsim 320\text{ }\mu\text{eV}$) although the difference in the lifetimes of the quasiparticles created in the top and bottom films will favour those created in the larger gap film. It is assumed that the probability for phonons, with energies above the Al energy gap, being absorbed is near one for the thicknesses of films employed.⁹⁴

⁹³Al–Ox–Al junction capacitance is 6 $\mu\text{F cm}^{-2}$.

⁹⁴The m.f.p. of 2Δ phonons in Al films has been determined experimentally by Long [60, 101] to be $\Lambda_L \approx 1600\text{ }\text{\AA}$ and $\Lambda_T \approx 3400\text{ }\text{\AA}$.

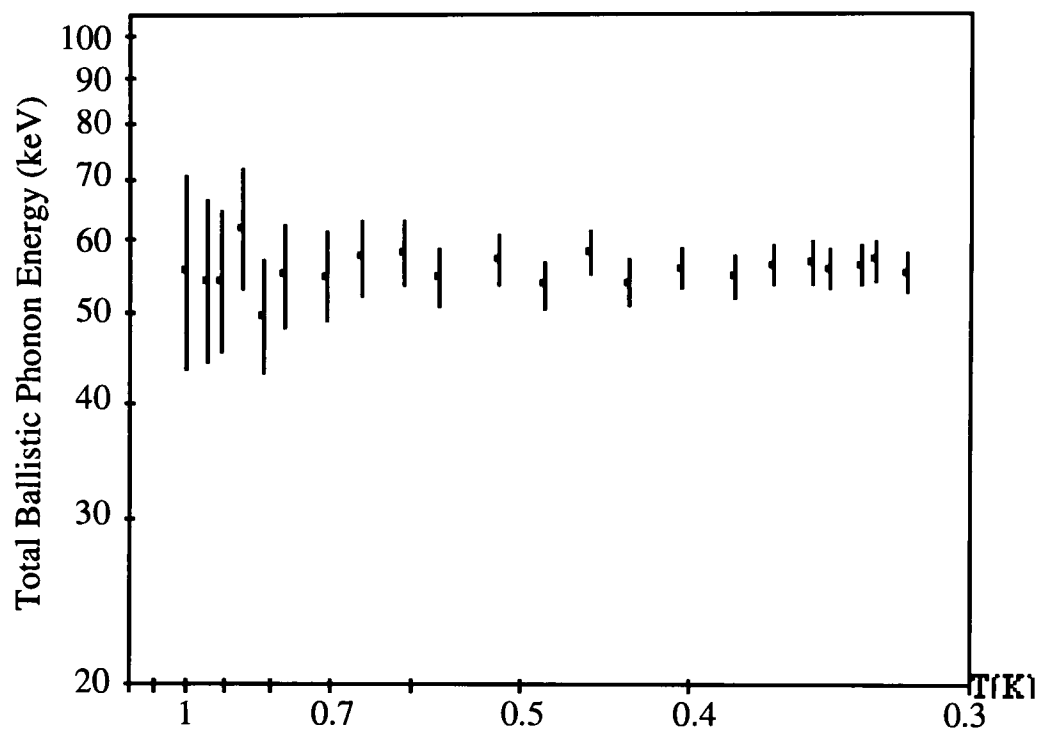


Figure 5.5.2 Ballistic Phonon flux versus temperature. The temperature dependence of the Al STJ has been adjusted for. The data are consistent with no temperature dependent attenuation of ballistic phonons below 1 K. The temperature axis is scaled as T^{-1} .⁹⁵

Figure 5.5.2 shows the variation in the time integrated L and T ballistic phonon flux (2–6 μ s) in the temperature range 0.3–1.0 K following laser pulse illumination of the far face of the crystal. In order to calculate the phonon flux from the voltage signal of the STJ it was necessary to adjust for the dynamic resistance of the STJ and the recombination time of the quasiparticles in the Al films⁹⁶. The STJ was current biased at the position of maximum dynamic resistance on the IV characteristic between $\Delta_{\text{Al}} - 2\Delta_{\text{Al}}$ and the dynamic resistance was measured directly from the IV characteristic. The Al recombination time was measured from the decay time of a STJ voltage pulse following direct illumination of the junction by laser light. The behaviour of the quasiparticle recombination time in the STJ is discussed in Appendix A. It should be mentioned here that the recombination time in the Al showed bias dependency at low temperatures implying that it was limited by self-heating in the junction. Within the experimental errors (arising, in the most part, from the uncertainty in the measurement of the Al recombination time) the ballistic phonon flux appeared constant in the range 0.3–1.0 K.

⁹⁵File: “nb140793 VIIIp71 Pulse Series”

⁹⁶This is discussed in detail in §3.4. In order to recover the ballistic features which occur over time-scales comparable to the measured recombination time in the Al (below 600 mK) Fourier transform techniques were employed.

The STJ (300 x 160 μm) subtends an angle from the point of interaction of 5×10^{-4} sterads. The integrated ballistic flux of order 50 keV would imply a source strength of ~ 1 GeV into 2π sterads. The estimated energy of the source (calculated from the temperature rise of the crystal, see §5.5.4) is 18 GeV.⁹⁷

Figure 5.5.3 shows the variation in the pulse shape (0-300 μs), for temperatures 0.3–1.0 K, after the deconvolution of the detector response function. The constancy of the ballistic feature at the leading edge can be seen. At 0.317 K the signal reaches a maximum at around 25 μs , shortly after the ballistic peak. It then decays with a time constant of 145 μs . At 1.00 K the signal rises immediately after the ballistic signal with a time constant of 36 μs . Figure 5.5.4 shows the pulses on a semi-logarithmic plot in the range 0–1.6 ms⁹⁸. At longer time-scales all the traces converge to a constant decay time of 10 ± 1 ms.

The parameters used to determine the absolute flux calibrations shown in Figs. 5.5.2–5.5.4, using the theory detailed in §3, are given in Table 5.5.1.

Parameter	Value	Comment
$2\Delta_{\text{film}}$	400 μeV	Average value
Quasiparticle recombination time, τ_r	100 ns–2 μs	Observed Values (see Appendix A)
Dynamic Resistance, R_D	0.8–8 Ω	
Quasiparticle tunneling rate, Γ_{tun}	$[2.8 \mu\text{s}]^{-1}$	Calculated using Eq. 3.4.4 and values below
$N(0)$ Al	12.2×10^{21} states eV cm ⁻³	[36]
Area	300 x 160 μm	
Film thickness	2500 Å	
Normal Resistance, R_{nn}	30 m Ω	

Table 5.5.1 Characteristics of Al–Ox–Al STJ.

⁹⁷ The ballistic flux deficit arises in part due to an overestimation of the STJ phonon detection efficiency but could also arise due to mechanisms within the Nb such as focusing, details of the initial phonon spectrum and phonon/quasiparticle yield immediately after the interaction. These points are discussed later.
⁹⁸ The original voltage signals were taken with a head amplifier with an AC-coupling time of 6.0 ms. This effect was folded out during the digital post-processing of the signal.

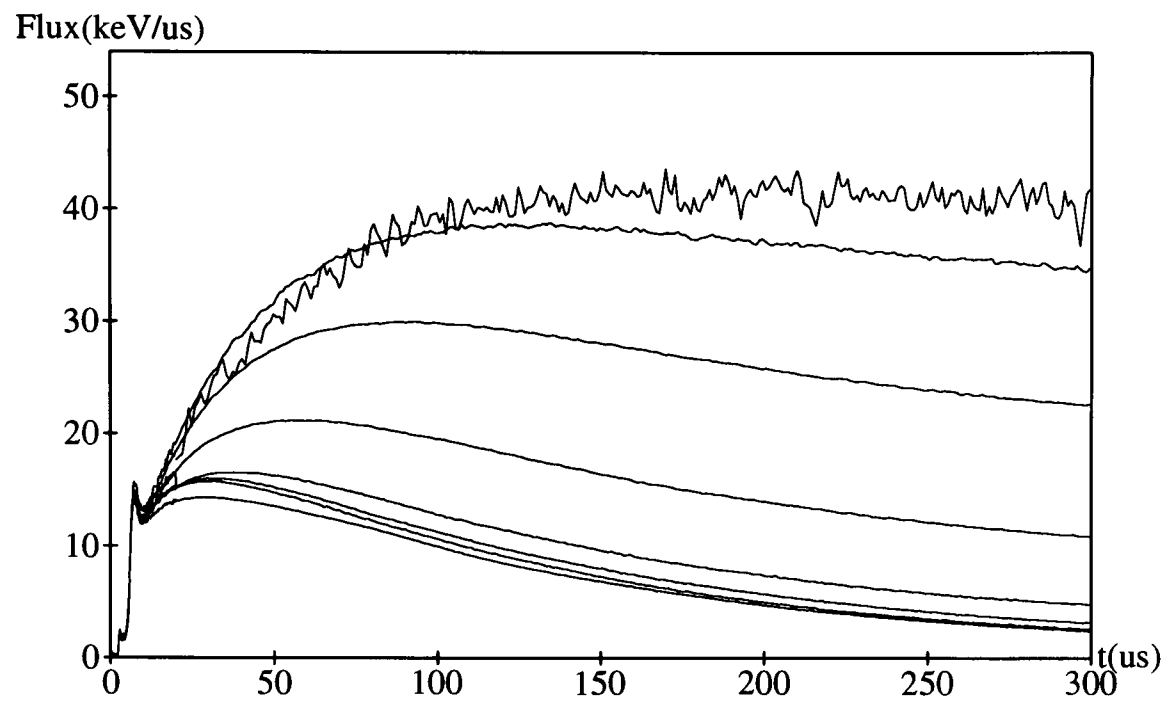


Figure 5.5.3 Phonon flux (in units of $\text{keV}\mu\text{s}^{-1}$) entering STJ, versus the time from the laser excitation, for temperatures, 0.317, 0.343, 0.380, 0.453, 0.571, 0.705, 0.858 and 1.00 K (curves run from bottom to top).⁹⁹

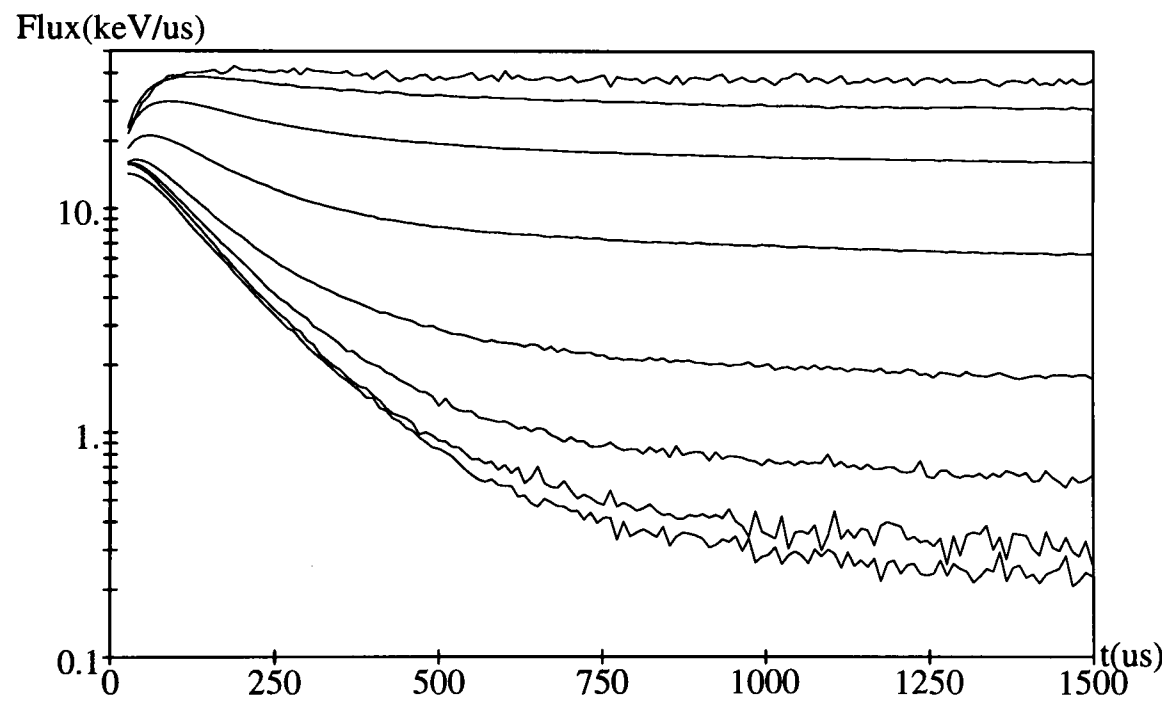


Figure 5.5.4 Log. plot of the phonon fluxes shown in Fig. 5.5.3.

We will discuss the pulses shape in more detail following our introduction of results from a Sn–Ox–Sn STJ in a similar geometry to this device.

⁹⁹File: “nb140792 VIIIp71 Pulse Series”

5.5.3. Detection using Tin Superconducting Tunnel Junctions

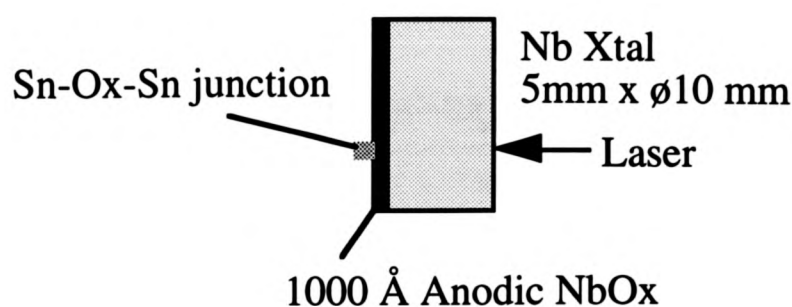


Figure 5.5.5 Geometry of Sn–Ox–Sn STJ on Nb crystal.

We studied the shape and temperature dependence of the non-equilibrium signal, arising from the laser excitation, at a different phonon energy threshold ($\Omega \gtrsim 1200 \mu\text{eV}$) from that of the Al STJs ($\Omega \gtrsim 400 \mu\text{eV}$). A Sn–Ox–Sn STJ was deposited on the insulated¹⁰⁰ surface of a 5 mm (ϕ 10 mm) Nb single crystal. The STJ will be sensitive to phonons above $2\Delta_{\text{Sn}}$ ¹⁰¹. In our experiment the Sn film thicknesses were 3000 Å, with top and bottom films in the same geometry as that for the Al–Ox–Al STJ of the previous section. At this threshold we calculate that the phonons will have a lifetime arising from anharmonic decay of 123 μs .

The leading edge of the signal in the STJ, following the laser excitation, was observed at $2.0 \mu\text{s} \pm 0.2$. This rules out the detection of any ballistic longitudinal phonons which would have an arrival time of 1.0 μs over 5mm. The leading edge time could indicate the arrival of ballistic FT phonons (1.9 μs), however, the shape of the leading edge was more consistent with diffusive transport. Within the constraints of signal to noise there was no peak in the deconvoluted pulse which would be associated with the ballistic propagation of the FT or ST (2.75 μs) phonons modes.

The deconvoluted pulse shape at 360 mK, which peaks at around 9 μs , was fitted using the diffusion model (see §3.5) and is shown in Fig. 5.5.6. The diffusion coefficient, which is determined by a fit to the leading edge of the pulse, was $0.5 \text{ m}^2\text{s}^{-1}$, which corresponds to a m.f.p for the L phonons of 300 μm . Assuming that the L phonon

¹⁰⁰It proved extremely difficult to fabricate good Sn–Ox–Sn junctions on SiO. A comparable thickness of anodic Nb oxide was substituted (see §4.3.2). We will assume that this doesn't materially alter the results, although this premise still needs to be confirmed.

¹⁰¹The mfp of 2Δ phonons in normal Sn films is about 1000 Å [60]. For a superconductor this will be reduced by 1:1.57. This was confirmed experimentally by Narayanamurti [102].

m.f.p. $\sim \Omega^{-4}$ (arising from impurity scattering) this would give a m.f.p. of 2.4 cm for $\Omega = 400 \mu\text{eV}$, which is consistent with the observation of ballistic phonons for the Al STJ data.

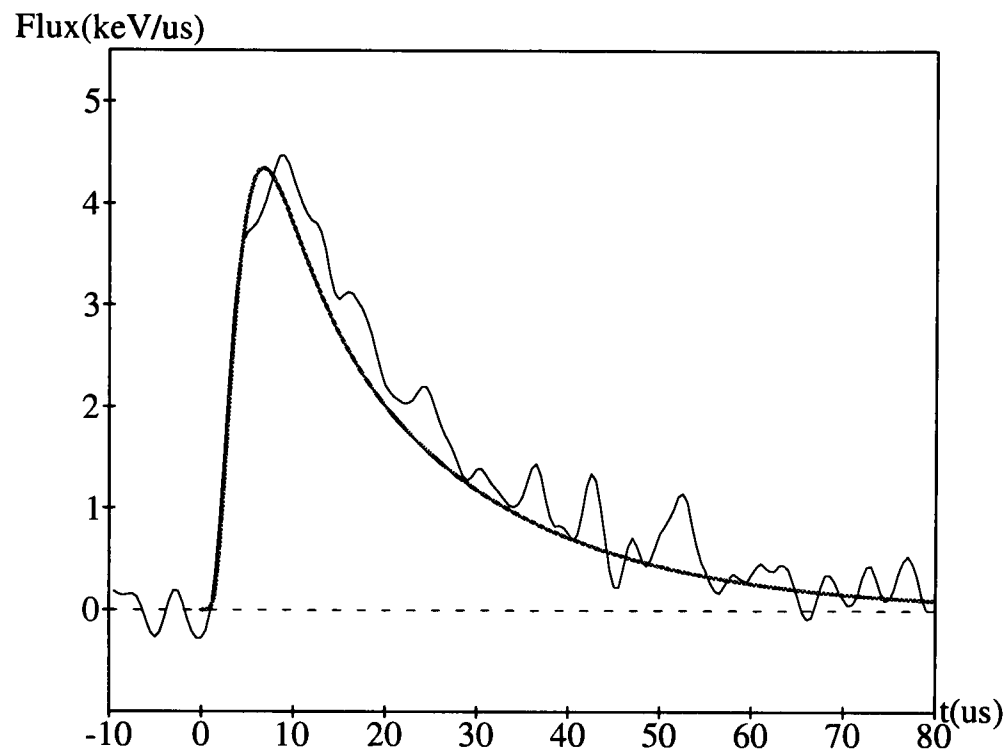


Figure 5.5.6 Signal in Nb crystal at 360 mK detected by Sn-Ox-Sn junction 5 mm from the laser interaction point. The STJ voltage signal was deconvoluted using $\tau_{RC}=1.0 \mu\text{s}$ and $\tau_r=5.4 \mu\text{s}$. The fit, shown as a grey line, was made using the 3-D diffusion model with $D=0.5 \text{ m}^2\text{s}^{-1}$ and a decay time of $20 \mu\text{s}$.¹⁰²

The phonon diffusion pulse is modified by the decay of the high energy phonons within the Nb. A value of $20 \mu\text{s}$ was found to fit the pulse adequately, which is lower than that estimated in §3.3.9 ($123 \mu\text{s}$). Beyond $60 \mu\text{s}$ an additional long lived tail¹⁰³ is present which cannot be due to above Sn gap phonons from the initial interaction, due to their short decay time. It will be discussed in the next section.

The change in the junction voltage signal shape with temperature is shown in Fig. 5.5.7 for 0.36-1.8 K. The signal amplitudes have been adjusted, by folding out the contributions of the dynamic resistance and the Sn recombination time to reflect the rate of energy entering the Sn films creating quasiparticles as was done for the Al junctions.

¹⁰²File: "nb051092 VIIIp104 Pulse Series"

¹⁰³The tail decay time, $3.5 \pm 0.3 \text{ ms}$, is independent of temperature in the range 0.8–2.0 K. We were unable to measure it accurately outside this range.

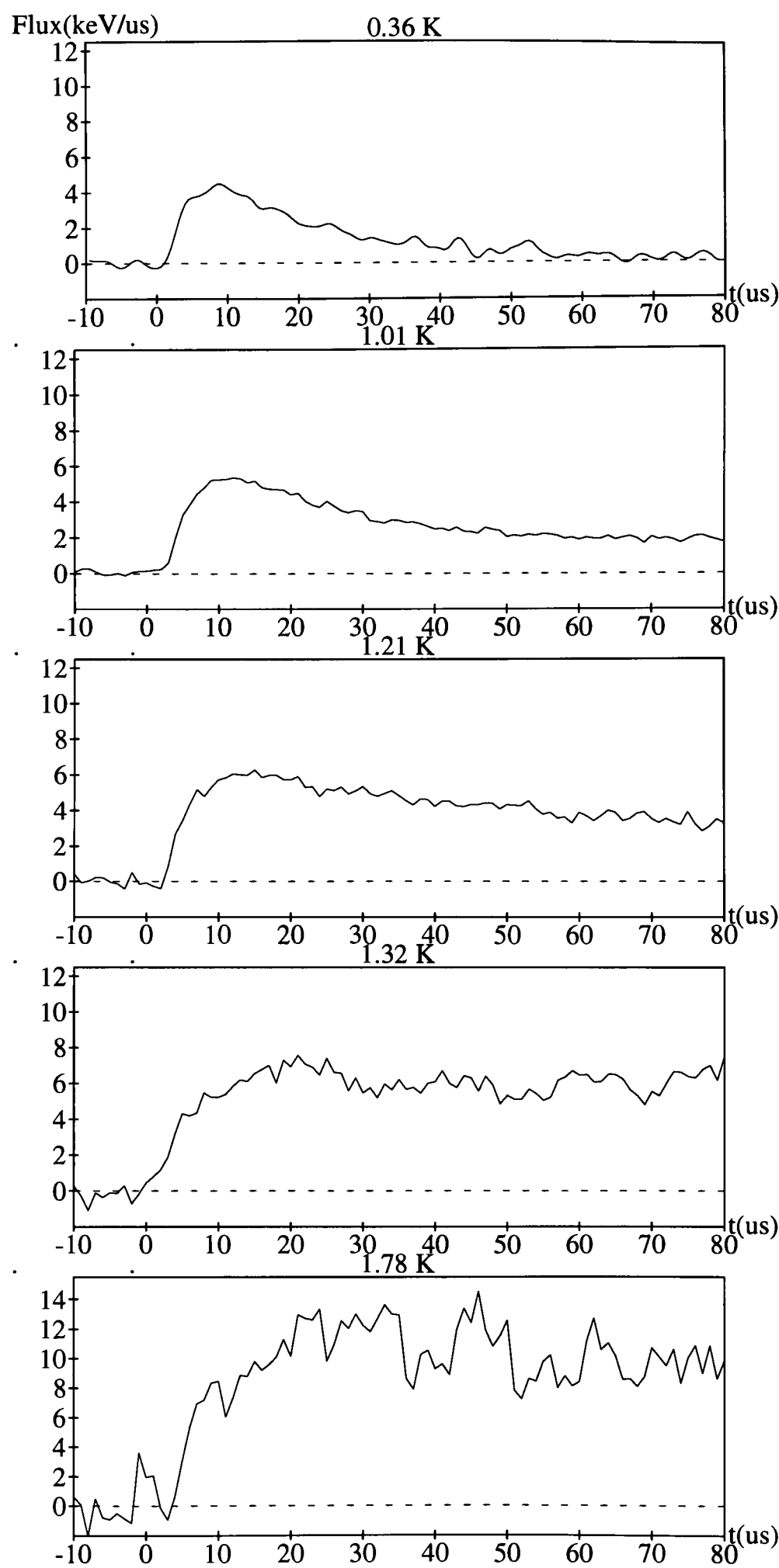


Figure 5.5.7 Temperature series (0.36 K-1.8 K) of pulses detected by Sn-Ox-Sn STJ on insulated surface of 5mm Nb crystal. Signal sizes are adjusted for the effects of the dynamic resistance and quasiparticle recombination time in the Sn films.¹⁰⁴

As would be expected the shape of the leading edge of the phonon diffusion pulse appears almost independent of temperature ($T \leq 1.8$ K) since the time for the scattering of

¹⁰⁴File: “nb051092 VIIIp104 Pulse Series”

phonons from quasiparticles, $\tau_{\phi qp} \gg \tau_{imp}^{ph}$, the impurity scattering time, at $\Omega = 1200 \mu\text{eV}$. A plot of the leading diffusion pulse height against temperature is shown in Fig. 5.5.8. In the region below 0.6 K the peak height is a constant. Above this temperature the peak maximum begins to increase. This is due to an increase in the contribution from the tail signal at time comparable with the diffusion peak. The superposition of the tail signal also moves the peak maximum to later times. This will be discussed shortly. Using the same techniques applied in the last section, the absolute flux calibration shown in Fig. 5.5.6–5.5.8 were made using the STJ parameterisation given in Table 5.5.2.

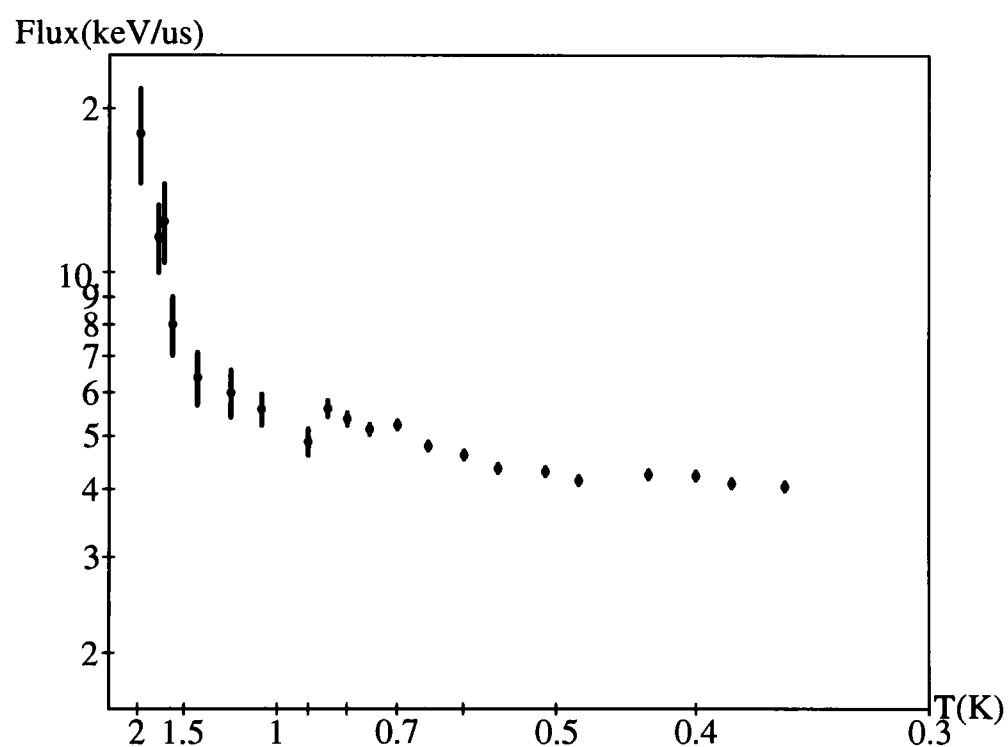


Figure 5.5.8 Phonon diffusion peak heights measured in energy flow into STJ per μs versus temperature for Sn-Ox-Sn STJ ¹⁰⁵.

Parameter	Value	Comment
$2\Delta_{\text{film}}$	1200 μeV	Average value
Quasiparticle recombination time, τ_r	30 ns–5.4 μs	See Appendix A
Dynamic Resistance, R_D	100 Ω	R_D fixed by parallel resistor
Quasiparticle tunneling rate, Γ_{tun}	$[525 \mu\text{s}]^{-1}$	Calculated using Eq. 3.4.4 and values below
$N(0)$ Al	8.14×10^{21} states eV cm ⁻³	[36]
Area	$300 \times 160 \mu\text{m}$	
Film thickness	3000 $\text{\AA}$	
Normal Resistance, R_{nn}	7 Ω	

Table 5.5.2 Characteristics of Sn–Ox–Sn STJ.

An estimate of the total phonon energy detected in the diffusion pulse (0–50 μs) at 0.360 K, is 100 keV. Since the junction covers 10^{-4} of the surface area of the crystal¹⁰⁶ this represents around 10% of our estimate of the initial energy deposited in the original interaction assuming the laser energy is ~ 10 GeV.

5.5.4. Signal Tail due to Quasi–Thermal Temperature Rise of Crystal

We still need to suggest possible mechanisms for a number of features, highlighted in the last two sections, in the pulses observed with the Al–Ox–Al and Sn–Ox–Sn STJs on Nb crystals.

Firstly, we need an explanation of the long lived tail signal which showed considerable variation in magnitude with temperature, when measured in terms of the creation rate of excess quasiparticles within the films. This is in contrast to the dominant ballistic and diffusion signals (of the Al and Sn devices, respectively), caused by phonons directly from the original excitation, which were independent of temperature measured in

¹⁰⁶Crystal Surface Area 5mm $\times$ ϕ 10 mm is 3.1 cm².

the same basis.¹⁰⁷ The tail signals were found to have a decay time, which was independent of temperature, although it varied for different experiments.

Secondly, we must identify the mechanisms giving rise to the time constants exhibited by the signals in the region 10–200 μ s. The signals either relax (at low temperatures) or rise (at high temperatures), to the tail signal level.

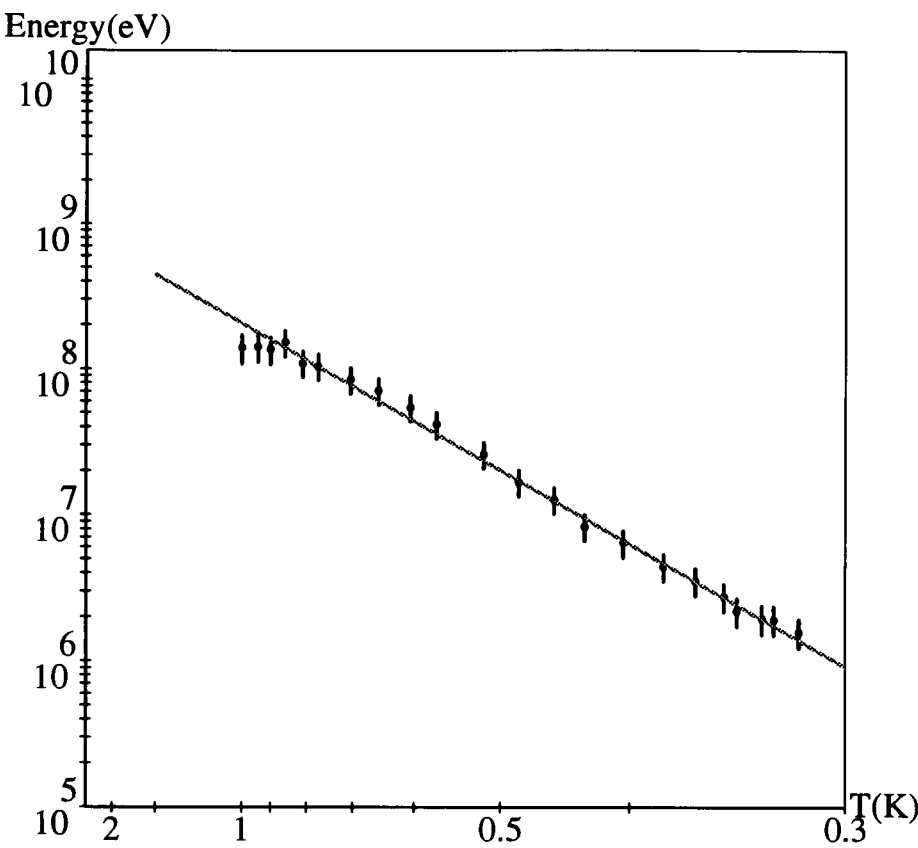


Figure 5.5.9 Temperature dependence of tail height (at 1.5 ms) for Al–Ox–Al device. Height is adjusted to reflect the energy required to create the number of non-equilibrium quasiparticles measured in the Al films (assuming a decay time of 10 ms). The line has a slope of 220 μ eV. The temperature is scaled as T^{-1} .

¹⁰⁷It should be remembered that the signals from the STJs were originally recorded as voltage pulses at constant current bias. These voltage signals typically varied by over three orders of magnitude in the pulse–temperature sequences shown in the previous sections. In analysing the pulse data we have converted the voltage signals to a measure of the rate of creation of excess quasiparticles in the STJ films in order to remove the nominal temperature dependence due to the nature of the STJ tunneling mechanism.

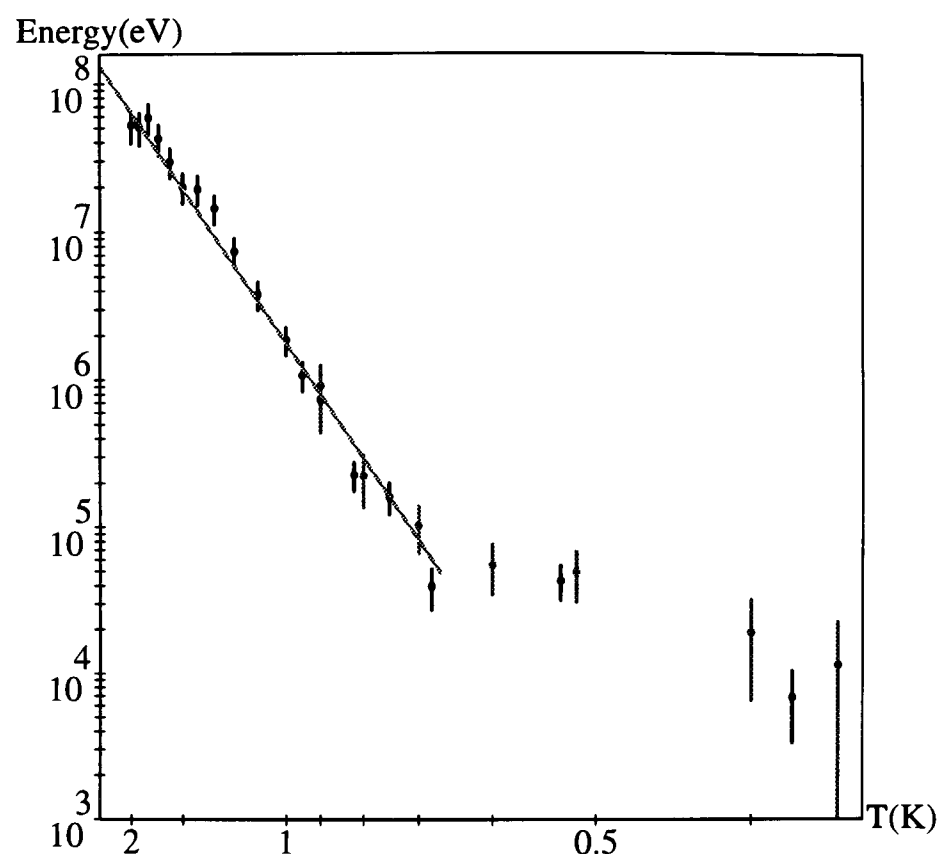


Figure 5.5.10 Temperature dependence of tail height (at 250 μ s) for Sn–Ox–Sn device. Height is adjusted to reflect the energy required to create the number of non-equilibrium quasiparticles measured in the Sn films (assuming a decay time of 3.5 ms). The line has a slope of 620 μ eV. The temperature is scaled as T^{-1} .¹⁰⁸

We will discuss the first of these effects. Fig 5.5.9 and 5.5.10 show the temperature variation in the apparent energy, required to create excess quasiparticles, for the duration of the long lived tail¹⁰⁹ in the in the Al and Sn STJs¹¹⁰. The signals appear to decrease as the temperature is lowered with slopes characteristic of the detecting film gap. Clearly, on the time scales considered here the total energy of the phonons entering the STJ films will be equal to that leaving. The number of quasiparticles within the films will be in near equilibrium with equal creation (due to 2Δ phonons) and annihilation (due to recombination) rates. It would, therefore, be more appropriate to consider the voltage signal of the STJ in terms of an apparent change in the temperature of the STJ film¹¹¹. The necessary calculations are shown in §3. The results are shown in Figs. 5.5.11 and 5.5.12.

¹⁰⁸File: "Pulse Temp VIIIp101 3.11.92"

¹⁰⁹This is the same as the flux parameterisation used in last sections except that we have integrated over the entire tail signal.

¹¹⁰The tail magnitude was measured at 250 μ s for the Sn STJ and 1.5 ms for the AL STJ to ensure that the contribution from initial phonon pulse is negligible.

¹¹¹We are making an assumption about possible changes in the phonon spectrum, within the STJ films, which will be justified at a later point.

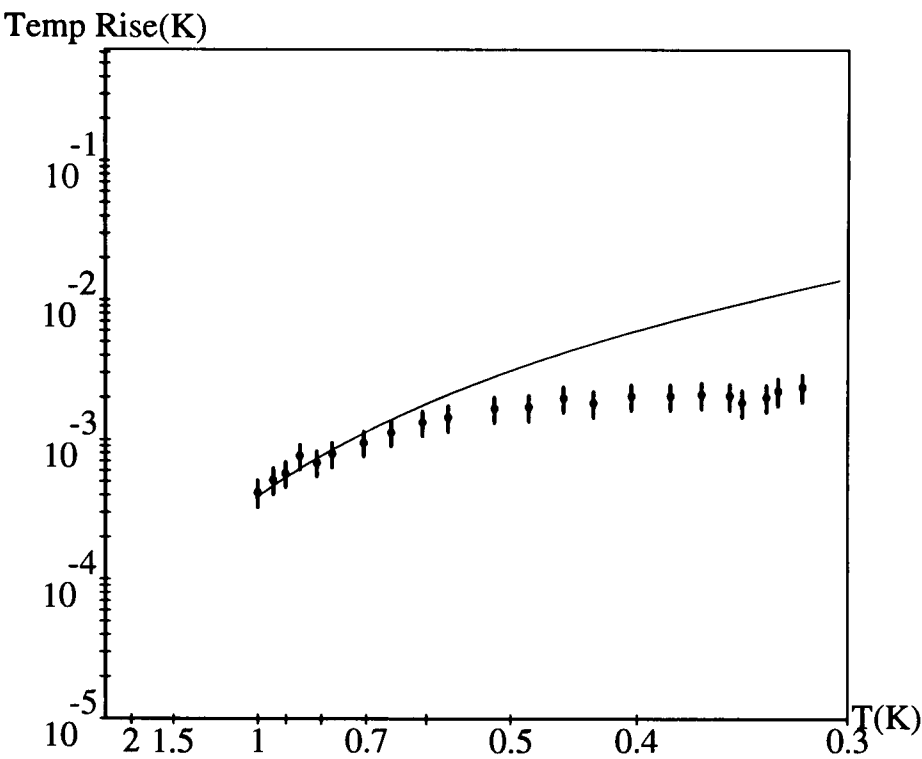


Figure 5.5.11 Temperature rise calculated from quasi-thermal equilibrium model of tunnelling current in Al–Ox–Al STJ plotted against the Nb crystal temperature prior to laser excitation. Line is expected temperature rise due to 18 GeV pulse energy deposition in the Nb crystal assuming a Debye lattice heat capacity. The divergence below 0.6 K is caused by STJ self-heating (see text for details).

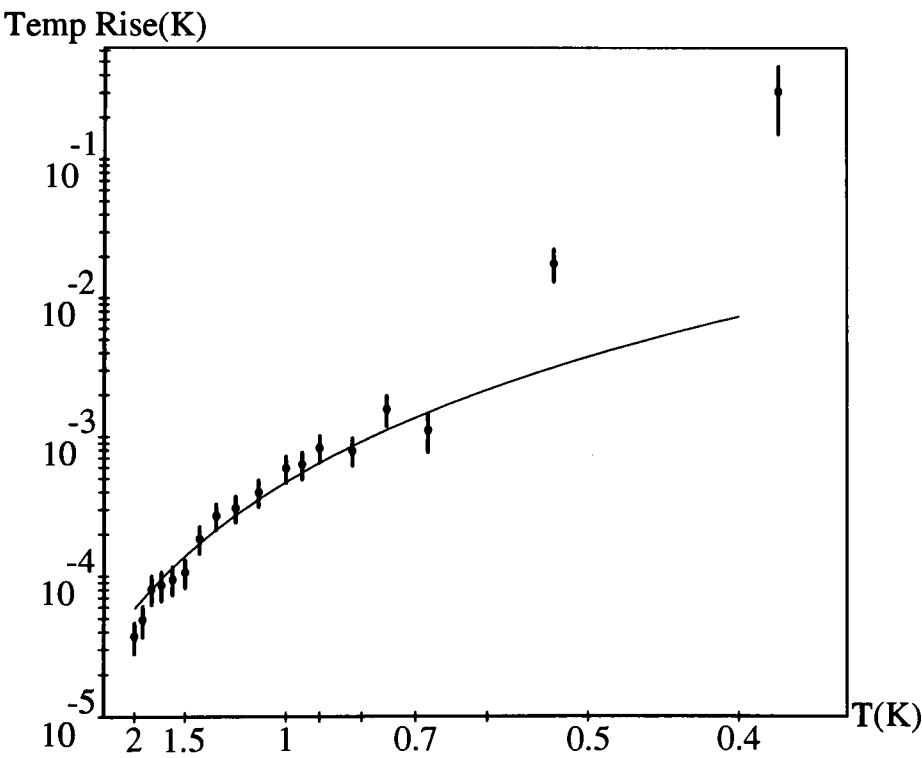


Figure 5.5.12 Temperature rise in Sn–Ox–Sn junction (calculated as Fig. 5.5.11). Line is expected temperature rise due to 10 GeV pulse energy deposition in the Nb crystal assuming a Debye lattice heat capacity. The energy deposited is the only free parameter in the fit and agrees with an estimate of the effective laser energy.

This approach may seem obvious, however, it is at odds with previous theoretical models and experimental observations of the behaviour of STJs on bulk crystals [103]. At

temperatures, well below the T_c of the STJ films, the tunneling current of a STJ is proportional to the number of phonons in the film above $2\Delta_{\text{film}}$. For an STJ on a ideal large single crystal it is generally assumed that phonons from an interaction will reverberate around the crystal. The above-gap phonons will be absorbed in the STJ films, creating quasiparticles. The quasiparticles will ultimately recombine in the films generating phonons at the gap energy. Many of these phonons will be retransmitted back into the bulk crystal and will be, subsequently, reabsorbed by the STJ film after traversing the crystal a number of times. The rate of creation of quasiparticles within the STJ films will therefore, to first order, be independent of temperature. The decay rate of the STJ signal would be determined by the anharmonic decay rate of phonons in the bulk crystal¹¹². Observations of the STJ signal behaviour in Nb are at odds with this interpretation. The amplitude of the tail signals, adjusted for the temperature dependence of the STJs, are not independent of temperature. The phonons, above the threshold of the STJ, are being thermalised on time scales much shorter than our estimates from the anharmonic processes in a single crystal.

We have calculated the expected temperature rise in the Nb crystals, on the basis that the initial energy from the laser is fully thermalised and that the Nb heat capacity is given by the Debye model ($c_v \sim T^3$). These estimates are compared to the temperature rise in the STJs implied by the variation in the excess voltage signals in Figs. 5.5.11 and 5.5.12. The absorbed laser energy has been adjusted to provide a best fit to the data. The values needed are very close to our own estimate of the laser energy per pulse assuming that the Nb surface is 95% reflective. The uncertainty in the laser fibre transmission coefficient and the reflectivity of the Nb surface make an absolute measurement of the heat capacity, by this method, difficult. However, the estimate of 10–20 GeV for the laser energy is consistent with the size of signals observed for α -particle (5 MeV) irradiation in similar geometries.

¹¹²The anharmonic decay rate of phonons increases as the temperature rises due to the boson enhancement factor arising from greater occupation of the decay states.

The data for the Al STJ, shown in Fig. 5.5.11, shows some divergence from the theoretical line below 0.6 K. This disagreement arises due to the self-heating in the high conductance junction. This was verified by observing the variation in the recombination time of quasiparticles within the Al film with bias at 0.360 K. The theory developed in §3.4.5 assumes that the quasiparticle population is that for the equilibrium temperature of the STJ and crystal. It will be demonstrated in the following section on Al SASTJs that the theoretical agreement is much better in high conductance Al-Ox-Al junctions when lower bias powers are used.

If we now review the pulses shown in Figs. 5.5.3 and 5.5.7 it is apparent that at higher temperatures ($T \gtrsim 1$ K) the signals in the Al and Sn STJs due to phonons from the initial interaction becomes dominated by a second signal which is evident only as a tail at low temperature ($T \lesssim 1$ K). As has already been detailed the size of the tail signal corresponds to that expected for a fully thermalised phonon distribution in the Nb crystal at a slightly higher temperature to that prior to the excitation. At all times the shape of the pulses will be determined by the population of phonons above the energy threshold of the STJ.

If we first consider the Al STJ pulse shapes. In a simple model of ballistic phonon propagation around the crystal, where the phonon number was unchanged after the initial excitation, the rate of phonons entering the junction at long time-scales would be about 30% of the integrated flux of phonons arriving ballistically at the STJ ¹¹³. This appears to over-estimate the level of the signal at, say, 25 μ s by about 20% for the pulse taken at 360 mK ¹¹⁴. The leading edge will also contain an additional component of higher energy phonons ($\Omega > 2\Delta_{\text{Al}}$) which diffuse from the interaction point due to their shorter m.f.p.'s. This will further complicate the structure of the first ~ 40 μ s of the signal ¹¹⁵. The subsequent decay of the STJ signal must arise from the net loss of phonons of energies

¹¹³The steady state flux would be given by the kinetic theory value of $1/4 Nc$ where N is the number density of phonons and c is the propagation velocity. The integrated ballistic flux is given by the solid angle subtended by the area of the junction from the original point of interaction.

¹¹⁴An estimate for the specular reflection coefficient of L phonons obtained in §5.2.1 was 50% although this is subject to large uncertainties.

¹¹⁵We have not applied our diffusion model to the leading edge since as yet it lacks the complexity necessary to handle both ballistic and diffusion components.

above the STJ threshold. At 0.360 K the decay time is 145 μs . This is over an order of magnitude shorter than that estimated from a calculation of the anharmonic decay time for L phonons in Nb. It seems likely that the down-conversion arises either in the Al films of the STJ or at the surfaces of the Nb crystal. Investigations of the down-conversion of phonons in other bulk crystals due to Al over-layers seem to indicate that they are not responsible for this effect [104]. Indeed, we have seen no increase in the phonon decay rates in Nb for samples covered in larger areas of Al. We believe the effect arises from inelastic decay and mode conversion of the phonons in the region of lattice distortion at the Nb surface. A 145 μs decay time in the 10 mm $\times$ $\varnothing$ 10 mm Nb crystal would imply a probability of down-conversion at the Nb surface of 0.03 assuming the signal was entirely due to ballistic phonons ($c = 2 \text{ km s}^{-1}$).

The pulse shape in the Al device at 1 K shows that the rate of quasiparticle creation in the Al films actually increases after the initial ballistic burst. The absence of this increase in the low temperature pulse shape indicates that the increase must arise from a change in the excitation phonon frequency distribution due to the presence of the thermal phonon distribution. Phonons that are initially above gap will still tend to decay. However, three phonon processes (at the crystal surface) will also tend to work in the other direction combining sub-gap phonons, from the non-equilibrium distribution, with sub-gap phonons from the thermal population, to produce new above gap phonons. Clearly the probability of this process occurring increases significantly as the mean energy of the population of thermal phonons increases. The phonon population spectra at 0.360 K, 1 K and 2 K are shown in Fig. 5.5.13 compared to the detection thresholds of the Al and Sn STJs.¹¹⁶

¹¹⁶The average energy of the populations is given by $2.70 kT$.

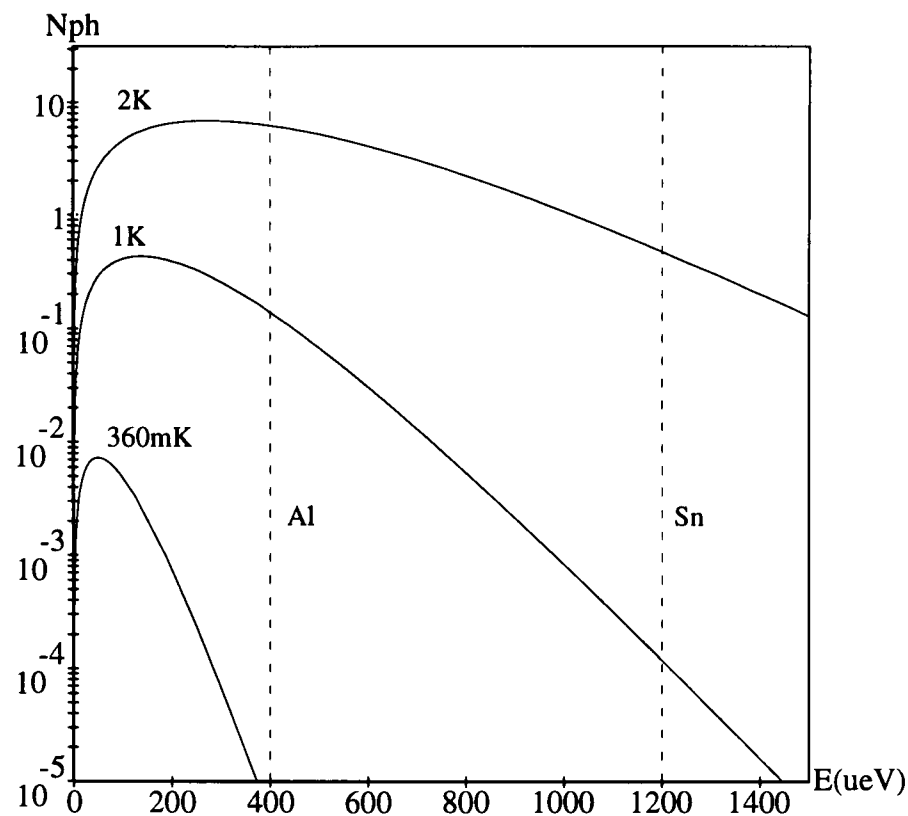


Figure 5.5.13 Phonon population against energy at 0.36, 1 and 2 K. The dashed lines show the threshold for detection with Al and Sn STJs.

The source of these non-equilibrium phonons, that only make a contribution to the signal after $\sim 10 \mu\text{s}$, could arise from two sources. The initial excitation may be generating a significant number of below $2\Delta_{\text{Al}}$ phonons that are subsequently up-converted as previously described. Alternatively the initial excitation may create a significant number of high energy phonons. Detection of these phonons will be highly inefficient for reasons illustrated in Fig. 3.4.2. For example those in the window $2\Delta_{\text{Al}} \leq \Omega < 4\Delta_{\text{Al}}$ will only break one Cooper Pair in the STJ ¹¹⁷. The energy of the phonon distribution will subsequently be redistributed by phonon up- and down-conversion giving rise to a possible additional signal in a manner similar to that mentioned in the first case. At present we have insufficient data to distinguish between these possible mechanisms.

The pulse shape from the Sn device at 0.360 K shows a decay time of $20 \mu\text{s}$. This is shorter than that estimated for the anharmonic decay time of L phonons in Nb although the discrepancy is not as large as that for the Al data. If it is assumed that the decay arises solely from surface effects, the apparent down-conversion probability is 0.24 calculated

¹¹⁷This will give rise to two quasiparticles, one at an energy Δ and the other at $\Omega - 2\Delta$. The latter will relax quickly to gap edge emitting a sub-gap phonon.

using our diffusion model with a diffusion coefficient from the fit in Fig. 5.5.6 of $0.5 \text{ m}^2\text{s}^{-1}$ and assuming a phonon velocity of $\sim 2 \text{ km s}^{-1}$.

5.5.6. Inelastic Scattering at Crystal Boundaries

We believe that the thermalisation of the non-equilibrium distribution of phonons generated by the laser is taking place at the surface of the Nb crystals. Experimental results from Wigmore [105] using GaAs bolometers on sapphire suggest that phonon boundary scattering is not only diffuse, but also involves at least partial absorption and thermalisation of the incident phonon distribution. At each reflection the re-emitted phonon distribution corresponds to that for a slightly lower temperature. He also finds that the thermalisation process appears to be independent of phonon frequency (in the range 1.9-5.0 K).

Worlock has proposed a model of the boundary scattering of phonons [106, 107]. He asserts that the phonons are not scattered by the visible surface of the material, as postulated by Casimir, but by a layer of dislocations $50 \text{ }\mu\text{m}$ thick, lying immediately below the surface. Thacher [107] reports results obtained on LiF at low temperatures which seems to support the importance of the structure of defects to a depth of $50 \text{ }\mu\text{m}$ below the surface and not just the surface condition. Sandblasted crystals at the lowest temperature were found to exhibit a boundary limited conductivity, λ , in line with Casimir's predicted relation, $\lambda \propto dT^3$, where d is the sample width and T is the temperature. Thacher confirms that the T^3 relation holds for temperatures below that where boundary scattering dominates (60 mK-6 K). Boundary scattering must therefore be a frequency independent phonon process except if it has the unusual functional form $f(\omega T^{-1})$.

Callaway's [108] theoretical analysis of phonon scattering processes divides them into two different types: N-processes (scattering which conserves crystal momentum) and U-processes (where crystal momentum is not conserved). The former will not lead to a finite thermal conductivity and creates a displaced Plank distribution. U-processes,

however, tend to an equilibrium Plank distribution. Callaway points out that boundary scattering is in the latter category.

Attempts have been made at the quantitative analysis of the boundary scattering of thermal distributions of phonons, however, the application of these techniques to non-equilibrium distributions presents a number of difficulties and will not be proceeded further here.

5.5.7. Detection using Aluminium Series Arrays of Superconducting Tunnel Junctions

The laser pulse experiment, detailed in §5.5.2, for an Al–Ox–Al STJ was repeated in a similar geometry using an Al SASTJ. The SASTJ was a series connected array of 150 Al–Ox–Al junctions fabricated on the SiO insulated surface of a Nb crystal, 15 mm x ø10 mm. The temperature dependence of the pulse shapes observed, in the range 0.3–1.0 K, were very similar to those for the Al–Ox–Al STJ. At 0.360 K the initial pulse was observed to decay with a time of $135 \pm 10 \mu\text{s}$ which is close to that of the Al STJ device at the same temperature. The current bias of the SASTJ was chosen so that the effective voltage bias point of each of the junctions was 10% of $2\Delta_{\text{Al}}$. This minimised any effects due to the self-heating of the individual junctions despite of their high conductance ($7.5 \times 10^{-6} \Omega\text{cm}^2$). The recombination time of the quasiparticles in the Al films was measured (using direct laser illumination) in the range 0.33–1.0 K and showed a characteristic slope of 220 μeV with no evidence of saturation at low temperatures. For a constant laser excitation the effective temperature rise of the SASTJ was calculated (using the theory in §3.4.7) from the tail signal voltage at 1.5 ms at a number of crystal equilibrium temperatures. The agreement with our estimate of the temperature rise based on the Debye heat capacity is extremely good and is shown in Fig. 5.5.14.

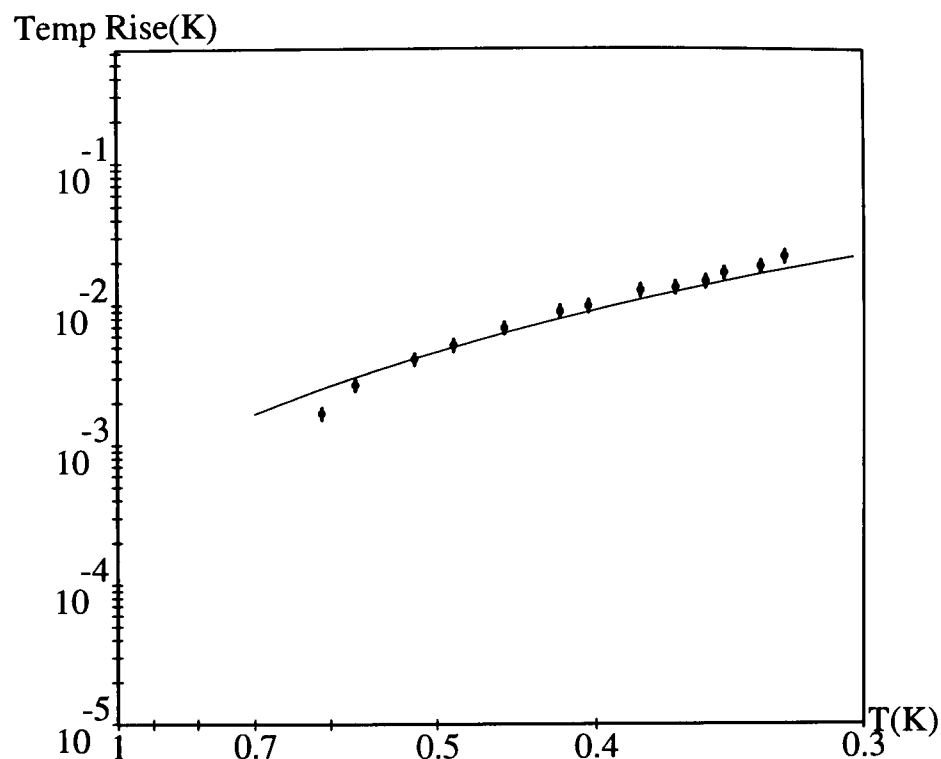


Figure 5.5.14 Temperature rise calculated from quasi-thermal equilibrium model of tunnelling current in Al–Ox–Al SASTJ plotted against the Nb crystal temperature prior to laser excitation. Line is expected temperature rise due to 30 GeV pulse energy deposition in the Nb crystal assuming a Debye lattice heat capacity. ¹¹⁸

The details of the electronic equivalent circuit represented by an SASTJ will not be discussed in detail here [109]. In summary, the SASTJ behaves electronically with a dynamic resistance, R_D , and capacitance, C ,¹¹⁹

$$R_D^{(Array)} = nR_D^{(Junction)}, \quad [5.5.1]$$

$$C^{(Array)} = \frac{C^{(Junction)}}{n}, \quad [5.5.2]$$

where n is the number of series connected junctions and R_D and C (on the left-side of the equations) are the dynamic resistance and capacitance of each junction. Thus the effective electronic RC time constant of the array is identical to that of a single junction. This is important since we are observing signals on time-scales ~ 100 ns. The SASTJ covers a much larger area of the Nb crystal than a single STJ leading to a proportionately larger

¹¹⁸File: "Plot Peak/Tail VIIIp66 9.7.92"

¹¹⁹The full form of the equations are $R_D^{(Array)} = \frac{(nR_D^{(Junction)})R_L}{R_L + (nR_D^{(Junction)})}$, and $C^{(Array)} = \frac{C^{(Junction)}}{n} + C_i$ where R_L is the

AC load-line given by the parallel combination of the bias resistance and amplifier input impedance, and C_i is the sum of the amplifier input capacitance and stray capacitance. The reduced form of the equations is valid in the limit that $R_L \gg R_D^{(Array)}$ and $C^{(Array)} \ll C_i$.

voltage signal from a uniform above-gap phonon flux (assuming the STJ and SASTJ have the same dynamic resistance).

We were able to check the enhanced sensitivity of the SASTJ by comparing results with the single Al–Ox–Al STJ in a similar geometry. The rate of energy entering a single junction of an SASTJ will be related to the observed voltage signal, δV , across the entire SASTJ by Eq. 1 in Table 3.4.4, where R_D becomes the dynamic resistance of the SASTJ¹²⁰. The calculated peak flux for a pulse arriving after laser excitation, observed at 0.360 K, using the parameters given in Table 5.5.3, was $20 \text{ keV}\mu\text{s}^{-1}$. The area of each junction in the SASTJ was 66% that of the in the Al–Ox–Al STJ (and the propagation distance was 50% further) and so the value compares favourably with the peak flux shown in Fig. 5.5.3.¹²¹

Parameter	Value	Comment
$2\Delta_{\text{film}}$	440 μeV	Sum of gaps divided by number of junctions
Quasiparticle recombination time, τ_r	100 ns–24 μs	See Appendix A
Dynamic Resistance, R_D	10–400 Ω	
Quasiparticle tunneling rate, Γ_{tun}	$[0.74 \mu\text{s}]^{-1}$	Calculated using Eq. 3.4.4 and values below
$N(0) \text{ Al}$	$12.2 \times 10^{21} \text{ states eV cm}^{-3}$	[36]
Area	$200 \times 160 \mu\text{m}$	
Film thickness	1250 $\text{\AA}$	
Normal Resistance, R_{nn}	3.5 Ω (23 m Ω)	150 junctions in series (Resistance per junction in brackets)

Table 5.5.3 Characteristics of Al–Ox–Al Series Array of Superconducting Tunnel Junctions (SASTJ).

¹²⁰Alternatively, the voltage signal across each junction of the array can be thought of as $\delta V/n$ across a resistance $R_D^{(\text{Array})}/n$.
¹²¹The apparent laser energy absorbed by the SASTJ from the temperature rise calculation was a factor of 2 higher than the Al–Ox–Al experiment.

5.5.7. Detection using Aluminium Series Arrays of Superconducting Tunnel Junctions

The decay times of the long lived tails in the signals from STJs and SASTJs on the Nb crystals will now be discussed in the context of an independent measurement of the thermal heat sinking of the Nb crystals.

The decay times did not appear to depend directly on the phonon energy threshold of superconducting films or the crystal temperature ¹²². The Sn-Ox-Sn data exhibited a tail with a decay time over an order of magnitude longer than our estimate of the anharmonic decay time of above threshold phonons. As has already been suggested the tail arises due to the sensitivity of STJs to changes in the above threshold component of thermal phonon distributions in the Nb (but with decreasing sensitivity as the temperature is lowered). If this is the case, the decay time constant will simply be given by the ratio of the heat capacity, C , and thermal conductivity of the heat sinking, K of the Nb crystals, ¹²³

$$\tau_{thermal} = \frac{C}{K}. \quad [5.6.1]$$

For temperatures, $T \lesssim 2$ K, the heat capacity of superconducting Nb is dominated by the phonon contribution which will be given by the Debye approximation,

$$c_v^{(phonon)} = 1945 \left(\frac{T}{\Theta_D} \right)^3 \text{ JK}^{-1} \text{ mol}^{-1}. \quad [5.6.2]$$

The theoretical phonon and electronic (normal and superconducting) heat capacities of Nb are shown in Fig. 5.6.1.

The thermal conductivity of the heat sinking of our Nb crystals is more difficult to estimate. In general the crystals were clamped into OFHC Cu blocks machined to approximately fit their cylindrical profile. GE varnish and a layer of cigarette paper was used to provide electrical isolation of the crystal from the sample holder. The crystal was firmly clamped into the Cu block, however, given the difficulty of matching the

¹²²The times scale (approximately) with the crystal volume.

¹²³ C is in energy per unit temperature and K is power per unit temperature.

deviations of the crystal from the profile of the clamp it is difficult to estimate the proportion of the crystal directly in contact with the GE/paper sandwich. Additional GE varnish was used to take up some of the slop.

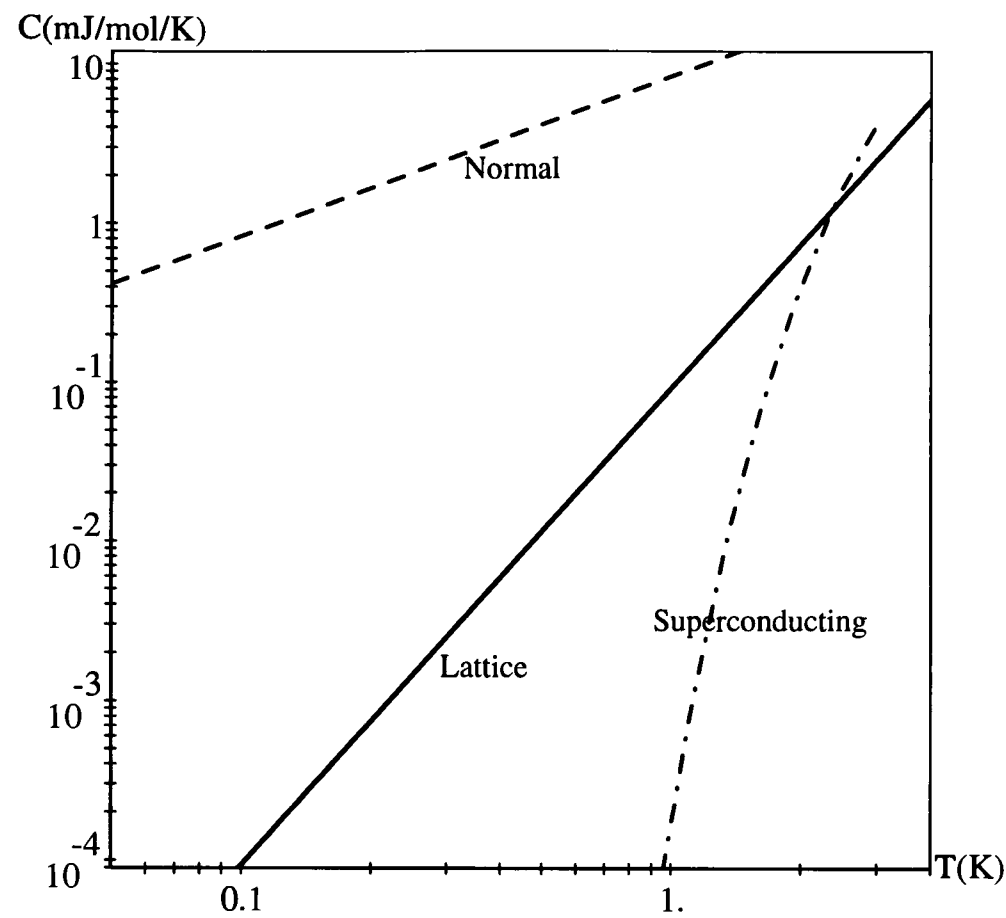


Figure 5.6.1 The lattice phonon and electron (in normal and superconducting state) components of the heat capacity of Nb in $\text{mJ mol}^{-1} \text{K}^{-1}$.¹²⁴

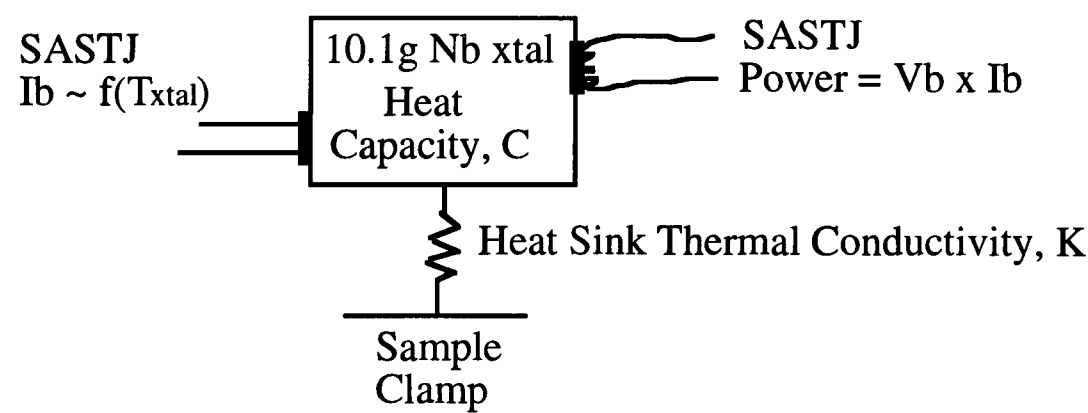


Figure 5.6.2 Schematic representation of a Nb crystal during an experiment to measure the thermal conductivity of the heat sinking. Heat was dissipated in one SASTJ. The temperature of the crystal was calculated from the variation in bias current of a second SASTJ.

An estimate of the heat sink thermal conductivity could be made by introducing a constant heater power into the Nb crystal and measuring the temperature rise. This was

¹²⁴ File: "Specific Heat Capacities 15.8.92"

achieved on devices with more than one SASTJ fabricated on the surface. The crystal is heated directly using one SASTJ which is biased using a current supply and the voltage drop across it measured four terminally. A second SASTJ on the opposite face of the crystal was then monitored for the corresponding change in bias current at a constant voltage. The temperature dependence of the bias current in the second SASTJ will follow Eq. 3.3.13 and was checked using a heater on the sample stage. The dependence of the thermal current versus stage temperature is shown in Fig. 5.6.3.

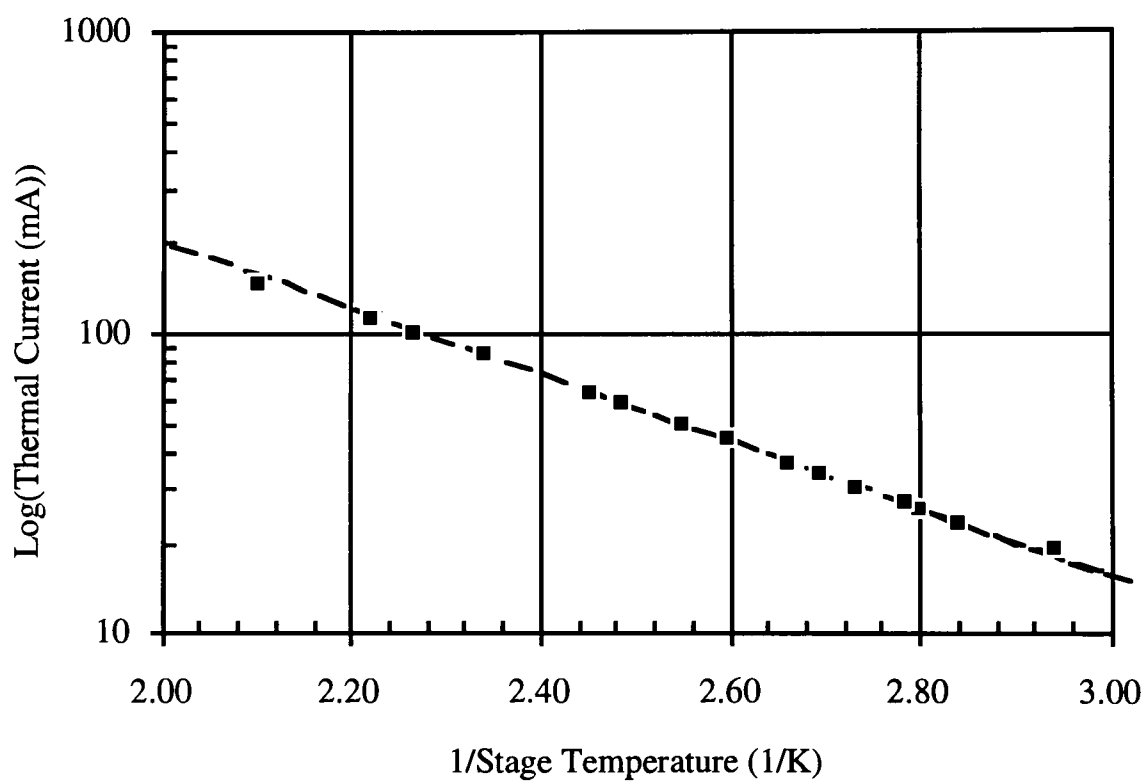


Figure 5.6.3 Variation in thermal current in SASTJ with temperature. ¹²⁵

A plot of the power dissipated in the first SASTJ verses the temperature rise in the crystal (above that of the sample stage) is shown in Fig 5.6.4.

The sample stage temperature was held constant using a separate heater mounted directly on it. Since the sample stage and crystal holder are constructed of Au plated OFHC Cu and tightly clamped together it was assumed they were at the same temperature.

¹²⁵File XL: "Al decay with bias (Series 1)" of nb020891 27/5/92 VIIp43

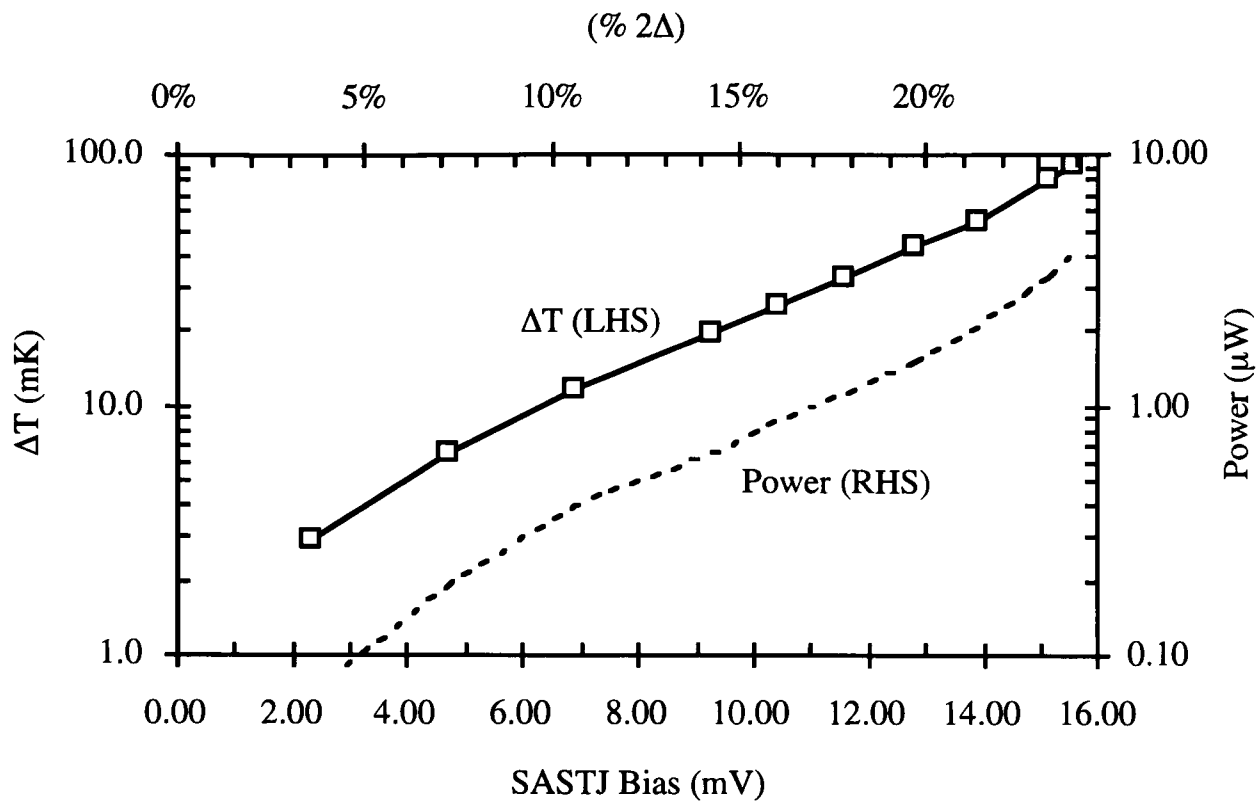


Figure 5.6.4 The temperature rise in a 10 g Nb crystal caused by the bias power of an SASTJ on its surface. The sample stage temperature was fixed at 360 mK. The SASTJ bias is shown in mV and as a percentage of 2Δ . Both the temperature rise of the crystal and the power dissipated in the SASTJ are shown.¹²⁶

The thermal heat sinking of the Nb crystal might be expected to be determined by the Kapitza resistance of those areas of the crystal touching the GE–Paper–GE–Cu crystal holder sandwich. Calculating the theoretical conductivity of such a composite would be invalidated by the uncertainties in the thermal characteristics of the materials and interfaces. We will, however, compare our results with those quoted for experiments on Nb-epoxy interfaces.

The thermal conductivity of the heat sinking is given by

$$K = \frac{\dot{Q}}{\Delta T}, \tag{5.6.3}$$

where $\dot{Q}$ is the power generated by the SASTJ, and ΔT is the temperature drop across the Nb crystal–holder interface. The Kapitza thermal boundary resistance, R_K , of this interface is then given by,

$$R_K = \frac{A \Delta T}{\dot{Q}}, \tag{5.6.4}$$

¹²⁶File XL: “Al decay with bias (Series 1)” of nb020891 27/5/92 VIIIp43

where A is the effective area of the interface. In the limit of small temperature differences the Kapitza resistance between two solids is expected to have a T^{-3} temperature dependence [110]. This dependence is derived from considering the balance between two equilibrium phonon distributions (temperatures T_1 and T_2) on either side of the interface. The net heat flow across the interface, area A , is found to depend on the temperatures of the phonon distributions as (Stefan–Boltzmann Law)

$$\frac{\dot{Q}}{A} = \alpha(T_1^4 - T_2^4), \quad [5.6.5]$$

where α is a constant determined by summation (over all phonon energies and modes) of the transmission of phonons at the interface. In the limit, $T_2 - T_1 = \Delta T \ll (T_1 \text{ and } T_2)$, and $T_1 \approx T_2 \approx T$,

$$R_K \approx \frac{1}{4\alpha} T^{-3}. \quad [5.6.6]$$

In analysing our data, where ΔT becomes comparable to the sample stage temperature, it is necessary to return to Eq. 5.6.5 and reexpress it in the form

$$R_K = \frac{1}{4\alpha} \left(\frac{T_1^4 - T_2^4}{4(T_1 - T_2)} \right)^{-1} = \frac{1}{4\alpha} (T^*)^{-3}. \quad [5.6.7]$$

where we relabel the complex temperature as T^* . Figure 5.6.5 shows $(R_K (T^*)^3)$ plotted versus ΔT for the Nb crystal with a sample stage temperature of 0.360 K. $R_K T^3$ is a constant ($= 0.0015 \text{ K}^4 \mu\text{W}^{-1}$) for temperature differences as large as 150 mK.

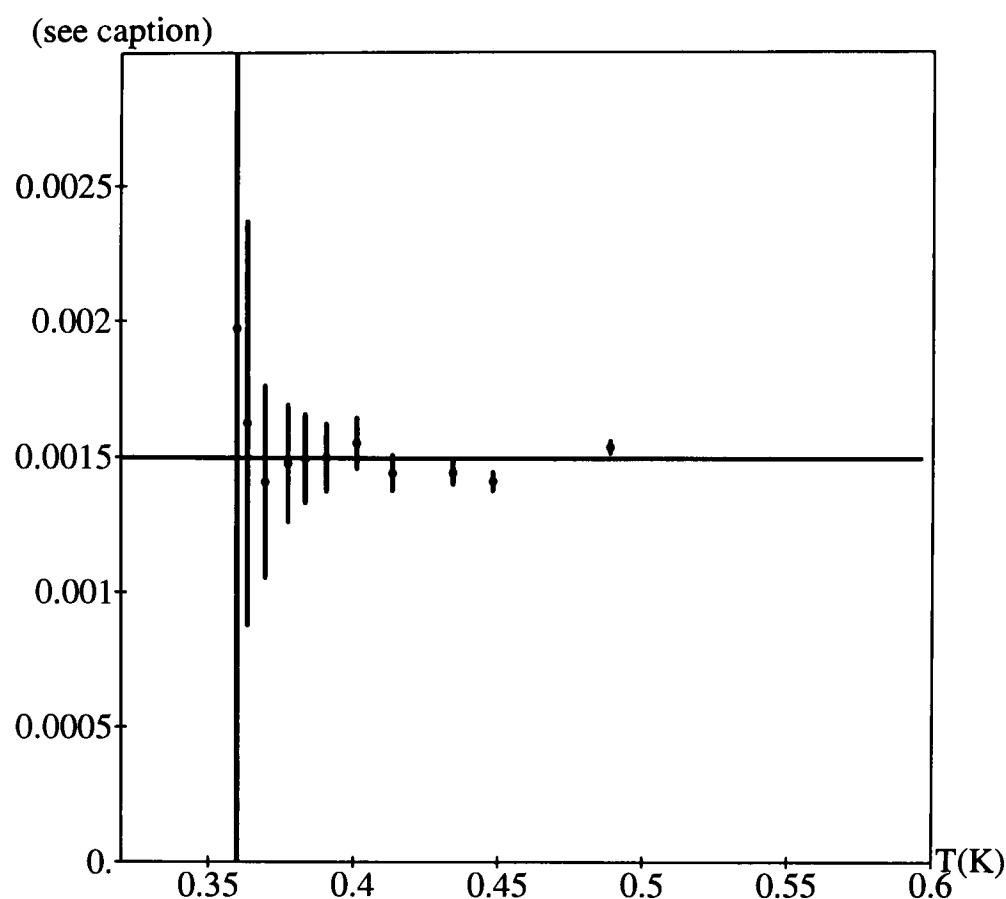


Figure 5.6.5 Plot of Kapitza Resistance function, $R_K(T^*)^3$ ($K^4\mu W^{-1}$) against the temperature of the Nb crystal. (See text for details of T^*).¹²⁷

A T^{-2} , rather than T^{-3} , dependence in the thermal boundary resistance has been observed for non crystalline/amorphous dielectric layers by Reynolds and Anderson [111]. They suggest that this temperature dependence may result from the interface showing some frequency dependence in its conductivity and acts like a low pass filter. This dependence was also observed by Schmidt for Nb-epoxy interfaces [112] when the Nb is in both the superconducting and normal states. Schmidt showed that at 1 K the ratio between the acoustic mismatch model prediction of R_K and his experimental results was a factor of 2 too high. He also showed that the ratio of Kapitza resistance for superconducting Nb to normal Nb was ~ 0.9 the reverse of theoretical estimates [113]. If we use Schmidt's value of R_K at 1 K ($\sim 10 \text{ cm}^2 \text{KW}^{-1}$)¹²⁸ we would have to assume that $< 0.1 \%$ of the surface area of our Nb crystal was contributing to the heat sinking. We estimate that the actual area in contact is around 10% and so conclude that the Ge-paper

¹²⁷File: "Kapitza Plot in Math 13.12.92" based on nb020891 Day270592 VIIIp43

¹²⁸Values for the Kapitza resistance of this order were measured for other superconductor epoxy boundaries [114]. However, it should be born in mind that the precise theoretical treatment of Kapitza boundaries with superconductors is complex (see e.g. [115]).

sandwich has a thermal resistance some two orders of magnitude worse than the systems studied by Schmidt.

At this stage of the development of the Nb single crystal detectors we have not been concerned with the quality of the thermal heat sinking. At low temperatures the response of STJs to thermal signals is vanishing small. The heat sinking is, however, important to the extent that poor sinking limits the bias power one can put into the SASTJs on the surface of the crystal when operating them to detect particles. Optimal voltage bias for the SASTJs appears to be around 10% of the 2Δ sum and since the bias currents decrease exponentially as the temperature is lowered it is possible to operate arrays of individual junctions with $m\Omega$ normal state resistances without causing self heating. This is the case even with the poor heat sinking calculated above. In cases where the Nb crystals were only loosely clamped the SASTJ performance was adversely affected by self-heating.

Using the value for the thermal conductivity of the heat sinking calculated above we derive a thermal time constant for the crystal of mass 10.1 g of ¹²⁹

$$\tau_{Thermal} = \frac{C}{K} = 15 \text{ ms.} \quad [5.6.8]$$

This is in close agreement to that experimentally observed using DC amplification of the STJ signal arising from the laser ($14.8 \pm 0.5 \text{ ms}$). ¹³⁰

For the Al–Ox–Al (§5.5.2) and Sn–Ox–Sn (§5.5.3) devices the long tail decay times were observed to be independent of temperature (0.3–1.8 K). This is consistent with heat capacity of the crystal varying as T^3 since it has been demonstrated that the heat sinking also has this temperature dependence.

¹²⁹ $c_v = 10.3T^3 \mu\text{JK}^{-1}$ using $\Theta_D = 275 \text{ K}$, mass = 10.1 g

¹³⁰File reference: VIIIp54

5.7. VARIATION OF SIGNAL WITH TYPE OF INTERACTION

5.7.1. Introduction

In principal, it is possible to vary the method of excitation of a superconductor in order to generate different phonon and quasiparticle frequency distributions. For example, in thin films, quasiparticles can be injected directly into the superconductor using a tunnel junction [45, 46].¹³¹ More suitable for investigation of transport phenomena in bulk crystals are microwave irradiation of the surface of the superconductor (photons of comparable energy to superconducting gap); the generation of phonons by STJs on the surface of the crystal [47, 116]; high energy photo-excitation of the crystal surface or the injection of phonons using thin film heaters. We were able to investigate the signals in bulk Nb from the last two sources (which will be discussed here), as well as investigating the signals arising from α -particle and γ -ray irradiation which will be covered in Chapter 7.

5.7.2. Phonon Generation by Thin Film Heater

Phonons were produced in a thin film heater, on the insulated surface of the Nb crystal. A resistive film can be heated using an applied current pulse, however, this method introduces considerable electrical pick-up into the cryostat which mask the leading edge of any non-equilibrium signal detected in a separate STJ. Alternatively, the thin film can be heated using laser excitation. (This was the method used in the experiments in this work.). The spectrum of phonons emitted from the heater will probably be characteristic of the black-body spectrum at the heater temperature, T_H , with a peak in phonon number at an energy of, $\Omega = 1.6 k_B T$. Weis [117] gives the form for the phonon number per unit area as

$$N(\Omega) = \frac{15}{\pi^4} \sigma \Omega^3 [n(\Omega, T_H) - n(\Omega, T_0)] \quad [5.7.1]$$

¹³¹The small number of quasiparticles injected into a superconductor by a biased tunnel junctions make them unsuitable for work with bulk crystals.

where $n(\Omega, T)$ is the Bose–Einstein distribution for the heater temperature, T_H and substrate temperature, T_0 . The Stefan acoustic constant is

$$\sigma = \frac{\pi^2 k_B^2}{120 h^3} \sum_{\lambda} \frac{\Gamma_{\lambda}}{v_{\lambda}^2}, \quad [5.7.2]$$

where Γ_{λ} is the acoustic-match ($0 < \Gamma_{\lambda} < 1$) of the phonon mode, λ , with velocity, v_{λ} , calculated for the interface of the film and substrate. A common problem in quantitative calculations for the heat pulse technique is the uncertainty in T_H . Pannetier [28] used a Cu film on the insulated surface of a Sn crystal in a very similar geometry to that we have used. He was able to estimate the proportion of phonons above the Sn superconducting gap (1200 μeV) by measuring the yield of quasiparticles that were created by pair breaking, immediately on the phonons leaving the heater and entering the Sn crystal. At the laser powers we were able to achieve in our experimental arrangement (0.1 W mm⁻²) his results indicated a negligible yield of above Sn gap phonons ($2\Delta k_B^{-1} = 13.9$ K). From this we can infer that the yield of phonons from our Cu film at the $2\Delta_{\text{Nb}}$ threshold (35 K) will be very small.

5.7.3. Photo-excitation of Superconductor Surface

We have also extensively studied the non-equilibrium signals arising in bulk Nb from direct irradiation by a GaAs laser ($\lambda \sim 900$ nm). The photon energy is comparable to the Fermi energy of the electrons in Nb and three orders of magnitude larger than the superconducting energy gap. The mechanisms of down-conversion of the energy initially deposited in a single free electron in a superconductor have been described by Chang and Scalapino [118]. Initially the electron will partition its energy between a number of other electrons through Coulomb interactions. The electron–electron scattering rate is,

$$\tau_{e-e}^{-1} \sim \frac{E_e^2}{\hbar E_F}. \quad [5.7.3]$$

After a number of collisions, the electron energy is low enough ($E < (E_F \Omega_D)^{1/2}$) for collisions involving phonons to dominate,

$$\tau_{e-\phi}^{-1} \sim \frac{\Omega_D}{\hbar} (E > \Omega_D) \text{ and } \sim E^3 (E \ll \Omega_D). \quad [5.7.4]$$

The relaxation phonons will themselves break more Cooper Pairs (if $\Omega > 2\Delta$) creating quasiparticles. Those phonons below the superconducting energy gap will be able to propagate without breaking Cooper Pairs. This cascade takes place extremely rapidly, on sub-ns time-scales. The yield of sub-gap phonons to quasiparticles from a photo-excitation is difficult to estimate theoretically and will be discussed further in §7. It is apparent, however, that the sub-gap phonon spectrum will favour higher energy phonons ($\sim \Omega^3$) unlike that from the black-body source ($\sim \Omega^2$) discussed in the previous section.

5.7.4. Experimental results

Figure 5.7.1 shows the time evolution of two signals, produced by (a) thin film heater and (b) photo-excitation, propagating 15 mm in the [110] direction of a Nb crystal at 0.7 K. The signals were detected using an Al SASTJ which is sensitive to phonons with energy greater than the Al energy gap ($\Omega \geq 2\Delta_{Al} = 340 \mu\text{eV}$) [103, 119].

The upper pulse in Fig. 5.7.1 was generated by a laser pulse incident on a 350 Å Cu film insulated from the crystal surface by 5000 Å of SiO [120]. As discussed in §5.8.2 the Cu film thermalises the excitation due to the laser pulse, thereby producing low energy phonons ($\Omega \ll 2\Delta_{Nb} = 3030 \mu\text{eV}$). The yield of quasiparticles in the Nb is expected to be extremely small. The sub-gap phonons will propagate ballistically through the Nb crystal, limited by impurity scattering. We estimate the threshold energy for an impurity m.f.p. comparable to the distance of propagation is 570 μeV for the T mode¹³². The structure in the signal in the first 15 μs arises from the differing speeds of propagation of the L, FT and ST phonon modes. The signal decays beyond 30 μs with a time characteristic of the thermalisation of the phonon population. This mechanism is discussed in §5.5.4.

¹³²This calculation is based on the impurity limited m.f.p. of phonons, in the high purity Nb, above the $2\Delta_{Sn}$ threshold determined in the experiment discussed in §5.5.3 and also assuming the m.f.p. scales as Ω^{-4} .

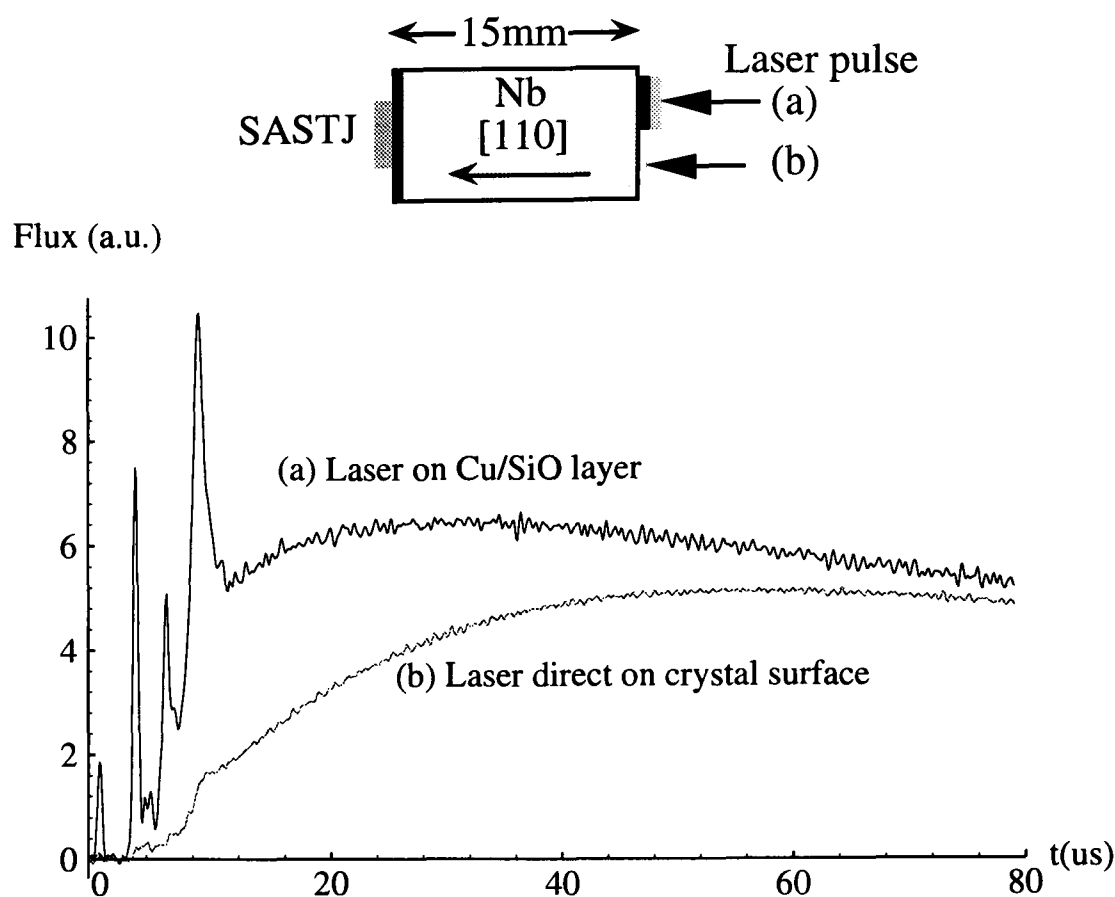


Figure 5.7.1 Signals due to laser excitation after propagating 15 mm in the [110] direction of a Nb crystal detected by Al SASTJ (Temperature 0.7 K). The signals are normalised on the tails at $> 200 \mu\text{s}$. The SASTJ response function has been deconvoluted from the signals (see §3.4.3). (a) Signal due to laser incident on a Cu/SiO layer on the surface of the crystal (black line) (b) Signal due to laser incident directly on the surface of the crystal (grey line).¹³³

The lower pulse (b) in Fig. 5.7.1 was generated by the direct photo-excitation of the crystal surface by the laser. The yield of ballistic phonon is seen to be considerably smaller than that for pulse (a).¹³⁴ The change in shape of the leading edge of the pulse arises because the average energy of the phonons produced in the initial interaction is much higher. The average impurity scattering m.f.p. of phonons is strongly energy dependent ($\sim \Omega^{-4}$) [121] leading to a more diffusion-like propagation for the phonon energy. The leading edge of the pulse is consistent with a diffusion coefficient of $1.4 \text{ m}^2\text{s}^{-1}$ if we assume the phonon spectra is unaltered during phonon collisions with the surface. We have already demonstrated that this is probably not the case. Figure 5.7.2 shows the significant alteration of simulated diffusion pulses shape (in the crystal geometry above) for a increase in the fraction of phonons being down-converted (below

¹³³File: "Ballistic Cross Xtal Vp102"

¹³⁴In a number of experiments using direct excitation of the crystal surface by the laser there was found to be some variation in the apparent yield of ballistic phonons. Compare Figs. 5.5.3 and 5.8.1(b). While some of this variation arises due to changes in the propagation distances of the signals the precise condition of the end of the laser fibre closest to the crystal and the Nb surface also factors appear to influence the ballistic yield. Residues left by poor cleaning of the illuminated Nb surface may be responsible. We intend to investigate this effect more closely in the near future.

the SASTJ threshold) at the surface from 0–0.24. It can be seen that the leading edge (and signal maximum) both occur at shorter times as the down–conversion factor increases. This type of analysis sheds some light on the factors effecting the shape of the phonon signals, however, the actual mechanisms will be considerably more complex. The spread in the spectrum of phonons from the initial interaction will lead to considerable variation in the phonon m.f.p.'s. When the phonons hit the crystal surface they will change frequency and mode altering their propagation characteristics. Any simulation of this behaviour will require a model of much greater complexity (and must cope with both diffusive and ballistic transport). A solution of a frequency and spatially dependent Boltzmann equation may provide such a model, but we have not pursued that technique here.

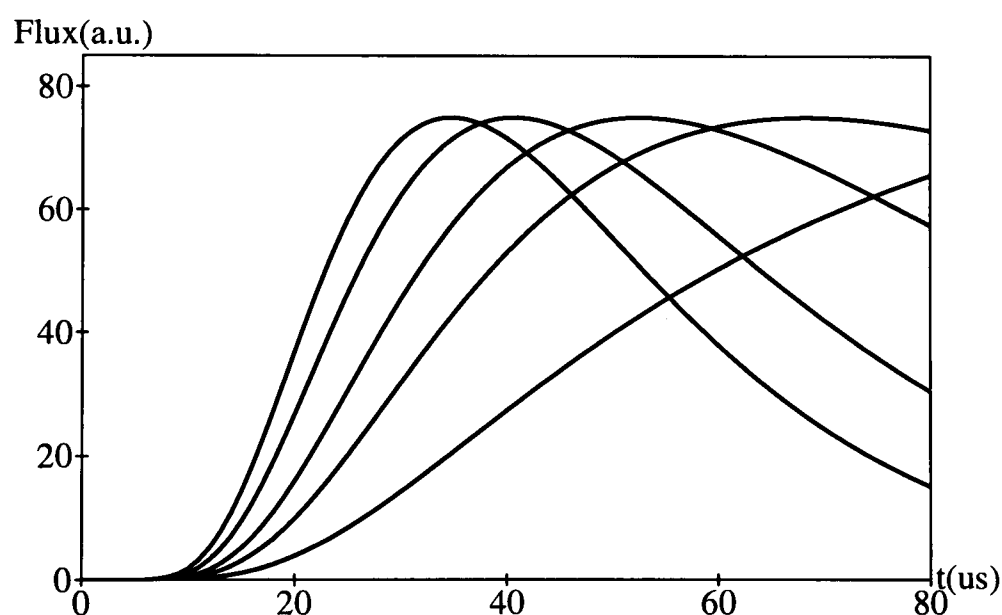


Figure 5.7.2 Simulation of phonon diffusion signal in a Nb crystal of the size given in Fig. 5.7.1. The phonons are assumed to propagate with a diffusion coefficient of $0.5 \text{ m}^2\text{s}^{-1}$. The effective down–conversion probability at the boundaries of the crystal has been varied producing a variation in the leading edge and peak times. The traces have been normalised to the same peak value. The curves running right to left have down–conversion probabilities of 0, 0.04, 0.08, 0.15 and 0.24.¹³⁵

The question of the contribution of quasiparticles to the signal also arises. In an experiment using methods of excitation similar to that shown in Fig. 5.7.1, Pannetier [28, 30, 120] was able to demonstrate an additional contribution to the non–equilibrium signal

¹³⁵File: “nb200591 Box Diff Sim 12.2.93”

in Sn crystals from quasiparticles.¹³⁶ The signals were detected by an Al bolometer which was insulated from the crystal. Such a sensor is not directly sensitive to quasiparticles but detects the phonons emitted by their recombination close to the bolometer.¹³⁷ The diffusion coefficient of the quasiparticles was high enough that they could be detected at times (~ 20 ns) well in advance of the arrival of the ballistic phonons (~ 500 ns). The same method of unambiguous identification is not available in the Nb since even in the highest purity sample the characteristic propagation time¹³⁸ is comparable to that of the phonons. We are continuing to investigate techniques for the simultaneous measurement of the quasiparticle and phonon components.

¹³⁶He was able to show that photo-excitation leads to a high yield of quasiparticles relative to ballistic phonons, whereas, a thin film heater (0.1 W mm^{-2}) produces low energy ballistic phonons and very few quasiparticles.

¹³⁷The size of the recombination phonon signal is strongly temperature dependent falling in proportion to the number of thermal quasiparticle in the Sn.

¹³⁸Quasiparticle diffusion coefficient is $0.7 \text{ m}^2\text{s}^{-1}$

6. PROXIMITY EFFECTS

In questions of science the authority of a thousand is not worth the humble reasoning of a single individual.

Galileo Galilei

6.1. INTRODUCTION

In order to study the structure of the superconducting energy gap in the Nb crystals STJs were fabricated directly on the surface of the samples. The STJs were formed with the Nb crystal as the base electrode, its surface was oxidised thermally (forming a tunneling barrier approximately 20 Å thick) and a second thin film superconductor (Pb) deposited using thermal evaporation (see §4). The current–voltage (IV) characteristic of such a junction is then given by the equation:

$$I(V) = \frac{1}{R_{nn}e} \int_{-\infty}^{+\infty} dE \rho_1(E) \rho_2(E - eV) [f(E) - f(E - eV)] \quad [6.1.1]$$

where $\rho_{1,2}(E)$ are the densities of states of the superconductor surface and probe film and $f(E)$ is the Fermi function. If the form of the superconducting density of states of the probe film is known the IV will yield information about the structure of the density of states of the Nb.

Shen [122] has pointed out that the electron-tunneling technique samples one coherence length¹³⁹ beyond the insulating oxide at low quasiparticle energies. In the case of transition metals, such as Nb, the low Fermi velocity leads to coherence lengths that are much shorter than those for sp-band materials, such as Pb. A contamination surface layer on a transition metal of $\sim 10\text{--}100\text{ \AA}$ may represent a substantial portion of the distance sampled in a tunneling experiment.

The details we obtain from the tunneling experiments will reflect the density of states of the bulk Nb as modified by the surface condition of the Nb single crystal. The results reported here for junctions directly on the Nb surface can be interpreted in the context of previous work on single crystal and polycrystalline Nb–Ox–Pb or Nb junctions (see Table 6.1 at the end of this chapter).

We have attempted to model the structure of the surface density of states of Nb in terms of a McMillan proximity layer [123]. The surface is assumed to be characterised by a thin region ($\sim 10\text{ \AA}$) of Nb that, due to damage from the ion implantation associated with ion milling during junction fabrication, is a normal metal. This approach was shown, by Keith and Leslie [124], to quantitatively predict the correct structure in the Nb–Ox–Pb IVs at around the sum of the energy gaps when compared to the expected ion implantation depth.

Nb–Ox–Pb (and Nb) junctions made by ourselves and others (listed in Table 6.1.1) show varying amounts of leakage current at biases between $\Delta_{\text{probe film}}$ and the sum of the energy gaps. Empirically this can be best explained by Nb (normal metal)–I–Pb tunneling taking place in parallel to Nb (superconducting)–I–Pb tunneling. This will be discussed in more detail later but it should be pointed out that, while both the alteration in the superconducting density of states and the additional normal state tunneling effects appear to arise due to surface contamination or damage of the Nb surface, it is evident that they

¹³⁹Coherence length, $\xi_0 \sim \frac{\hbar v_F}{\Delta_0}$ which for Nb, $v_F \sim 2 \times 10^5\text{ m s}^{-1}$ gives $\xi \sim 1000\text{ \AA}$. BCS theory predicts an additional factor of 0.32 in this expression.

Reference	$2\Delta_{\text{Nb}}$ (meV)	Comment
Sherrill and Edwards (1961) [125]	3.02	Polycrystalline films Nb–Ox–Pb (considerable variation in quality of the STJs).
Townsend and Sutton (1968) [126]	3.05 ± 0.05	Polycrystalline film Nb–Ox–Sn.
MacVicar and Rose (1968) [127]	3.10 ± 0.02	Single Crystal Nb–Ox–Pb.
Hafstrom et al. (1969)		Single Crystal Nb–Ox–Pb. Evidence for a second energy gap in Nb.
Frommer et al. (1973) [90]	3.12 ± 0.05	Single Crystal Nb–Ox–Pb. Absence of second gap in Nb tunnelling data. ¹⁴⁰
Bostock et al. (1976) [128]	3.12 ± 0.02	
Broom et al. (1975/6) [129, 130]	3.12	Polycrystalline film Nb-Ox-Nb/Pb. Includes an empirical fit of IV characteristics.
Wolf and Zasadzinski (1977/8) [131, 132]	3.02 ± 0.04	
Shen (1970) [133]	2.98	Polycrystalline film Nb-Ox-Nb
Keith and Leslie (1978) [124]	3.05 ± 0.05	Single crystal Nb-Ox-Pb. Correlated variation of shoulder structure in IVs with ion mill damage.
Gaitskell (1993) [this paper]	3.10 ± 0.05	Single crystal Nb–Ox–Pb

Table 6.1.1 Listing of tunnel junction experiments performed with Nb and the observed energy superconducting energy gaps (taken in part from Keith and Leslie [124]).

stem from different mechanisms. Different methods of surface preparation yield different amounts of each effect.

We have also investigated the behaviour of thin ($\sim 100 \text{ \AA}$) layers of Al which were deposited on the surface of Nb single crystals using our evaporation system. Attempts to explain the temperature variation of the induced energy gap in the Al proximity layers (at temperatures above and below the superconducting transition temperature, $T_c \sim 1.2 \text{ K}$, of

¹⁴⁰This paper corrected a series of previous papers, published by the Group at MIT, which misinterpreted low bias structure in the IVs of junctions on the surface of Nb crystals as evidence for a second superconducting gap in Nb.

Al thin films) using the McMillan model were only partially successful. This failure almost certainly arises due to the non-ideal nature of the Nb crystal surface (as mentioned above). While it may be possible to analyse the system using a multiple layer McMillan approach we have not proceeded with that idea in this work.

At the end of the chapter we discuss a single Al proximity layer (80–2000 Å) that were fabricated on Nb single crystals in our CVC sputtering system. This system is able to achieve oxygen partial pressures ($\sim 10^{-10}$ torr) over 100 times better than the evaporation system. A significant improvement in the degree of proximity effect was observed. Good agreement was found between the McMillan model and the temperature dependence of the induced Al energy gap.

We fabricated junctions directly on the Nb crystals in order to investigate the non-equilibrium behaviour of quasiparticles in the bulk Nb. Quasiparticles created by a laser or particle interaction in the Nb crystal propagate throughout the crystal. Those within a coherence length of the junction barrier will be able to tunnel giving rise to an excess current in the junction above the equilibrium thermal current. Additional experiments performed with quasiparticle trapping layers, formed by Al proximity films deposited in the evaporator, were inconclusive. The failure to observe quasiparticle trapping for Nb crystals with proximity films made in the evaporator may in some part arise from the contamination of the interface between the Nb crystal and the trap leading to extremely low transmission coefficients at this interface. Al proximity layers deposited in the CVC sputtering system showed much higher transmission coefficients at the Nb–Al interface but proved more difficult to fabricate junctions on. The crystals had to be removed from the CVC system and placed in the evaporator exposing them to the atmosphere. The normal state resistances of such devices were extremely high ($\gg 10$ k Ω) making their use for signal propagation measurements difficult. We intend to investigate devices made in this way in the future but cannot report any results of quasiparticle trapping at present.

We will now briefly summarise the McMillan model of weakly coupled superconductor–metal systems and then look at its application to the Nb_{bulk}–Nb_{surface}–Ox–Pb and Nb–Al_{proximity layer}–Ox–Pb geometries.

6.1.1. Brief Summary of McMillan Proximity Tunneling Model

McMillan [123] has considered the proximity sandwich of a superconductor and normal¹⁴¹ metal film, represented in Fig. 6.1.1, as a second–order perturbation problem where the tunneling of electrons across the S–N interface leads to coupling of the electronic wave functions of the two films. McMillan assumes that the thickness of S and N layers are thin compared to the m.f.p. modified coherence length. This avoids the problem of having to consider any variation in the superconducting order parameter. It is also assumed that the electrons undergo diffuse reflection at the boundary. The other assumptions are listed on p537 of McMillan [123] and relate to the simplification of calculations involving the electron tunneling matrix elements, T_{nn} , and the transmission probability of electrons, σ , through the interface. McMillan assumes the latter to be small.

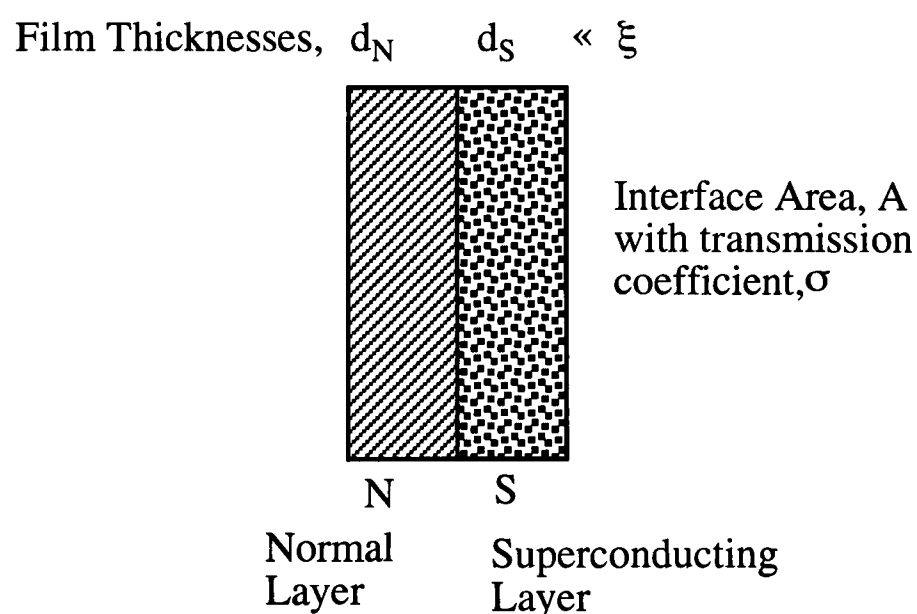


Figure 6.1.1 Superconductor—normal metal sandwich considered in McMillan's analysis.

¹⁴¹We will keep McMillan's original convention of referring to the second layer as a normal metal although this analysis can also be applied if the second film is a superconductor.

The equations of the model are developed in terms of the scattering rates of electrons in the two films, Γ_N and Γ_S which are given by¹⁴²

$$\begin{aligned}\Gamma_{N,S} &= \pi |T|^2 A d_{S,N} N_{S,N}(0) \\ &= \frac{\hbar}{2\tau_{N,S}}\end{aligned}\quad [6.1.2, 6.1.3]$$

where $N_{N,S}(0)$ is the electron density of states of one spin at the Fermi energy and $\tau_{N,S}$ are the electron relaxation times in the two films. Other definitions given in Fig. 6.1.1 above.

The relaxation times are given by

$$\tau_{N,S} = \frac{L_{N,S}}{v_{FN,S}\sigma} \quad [6.1.4, 6.1.5]$$

where $L_{N,S}$ are the average electron path length between collisions *with the interface barrier*, and $v_{FN,S}$ are the Fermi velocities. The average electron path lengths between collisions were shown by Vrba and Woods [135] to be given by

$$L_{N,S} = \left\{ 2d_{N,S} \right\} B_{N,S} \left(\frac{l_{N,S}}{d_{N,S}} \right) \quad [6.1.6, 6.1.7]$$

where B is a function of the ratio of electron mean free path, $l_{N,S}$, to film thickness. For a clean film they calculate $B = 2$. This gives

$$\Gamma_{N,S} = \frac{\hbar v_{FN,S}\sigma}{4B_{N,S}d_{S,N}} \quad [6.1.8, 6.1.9]$$

Using the above parameters McMillan shows that, for the proximity sandwich in which electrons are scattered between the two films, that the simultaneous self-energies $\Delta_N(E)$, $\Delta_S(E)$ for the fixed BCS potentials Δ_N^{ph} , Δ_S^{ph} are given by

$$\begin{aligned}\Delta_N(E) &= \left(\Delta_N^{ph} + \frac{i\Gamma_N \Delta_S(E)}{[E^2 - \Delta_S^2(E)]^{\frac{1}{2}}} \right) \left(1 + \frac{i\Gamma_N}{[E^2 - \Delta_S^2(E)]^{\frac{1}{2}}} \right)^{-1} \\ \Delta_S(E) &= \left(\Delta_S^{ph} + \frac{i\Gamma_S \Delta_N(E)}{[E^2 - \Delta_N^2(E)]^{\frac{1}{2}}} \right) \left(1 + \frac{i\Gamma_S}{[E^2 - \Delta_N^2(E)]^{\frac{1}{2}}} \right)^{-1}\end{aligned}\quad [6.1.10, 6.1.11]$$

¹⁴²In his original paper McMillan omitted the factors of p and 2 which was corrected by Kaiser and Zuckermann[134]. This original mistake lead to errors in some of the parameters quoted by later authors.

The self-consistency relations for the BCS potentials, at temperature T , are

$$\Delta_{N,S}^{ph} = \lambda_N \int_0^{\hbar\omega_{cN,S}} dE \operatorname{Re} \left(\frac{\Delta_{N,S}(E)}{[E^2 - \Delta_{N,S}^2(E)]^{\frac{1}{2}}} \right) \tanh\left(\frac{E}{2kT}\right) \quad [6.1.12, 6.1.13]$$

and the density of states of the excitations can then be found from

$$\rho_{N,S}(E) = \operatorname{Re} \left(\frac{|E|}{[E^2 - \Delta_{N,S}^2(E)]^{\frac{1}{2}}} \right) \quad [6.1.14, 6.1.15]$$

This model has been applied to a number of experimental situations with success even in those cases where the simplifying assumptions, in particular the limit on the film thickness, $d \ll \xi_0$, have not strictly been met. (see, for instance, Vrba and Woods [135] and Goldie [31]).

6.2. TUNNELING EXPERIMENTS ON THE SURFACE OF NIOBIUM CRYSTALS

6.2.1. Introduction

A number of attempts were made to fabricate STJs with a Nb single crystal as the base electrode. The crystals used ranged in RRR from 10^2 – 10^4 . No direct dependence of junction quality on crystal purity was observed. In all cases sample crystals were spark cut from the original boule and subsequently chemically polished prior to loading in the evaporation chamber (see §4). The period spent by the crystals in the atmosphere following chemical polishing was kept to a minimum (~15 minutes) but given the ease with which a clean Nb surface oxidises this exposure was enough to build a native oxide barrier. If no further cleaning and oxidation steps were taken prior to deposition of a Pb film the junctions were found to exhibit strong semiconductor-like behaviour¹⁴³ and have significant leakage currents.

¹⁴³The junction resistance increased by around 100 times on cooling from room temperature to the superconducting critical temperature.

In order to provide a clean surface for fabrication of the Nb–Ox–Pb junction the crystals were ion beam milled and then thermally reoxidised. Keith and Leslie^[124] have demonstrated that ion milling a polycrystalline Ta or Nb surface leads to a damage layer. They showed using Auger electron spectroscopy that the depth of the ion contamination corresponds to that estimated kinematically¹⁴⁴. For Ar atoms incident on a Nb surface the implantation depth of Ar is 6.5 Å per keV energy in the incident beam.

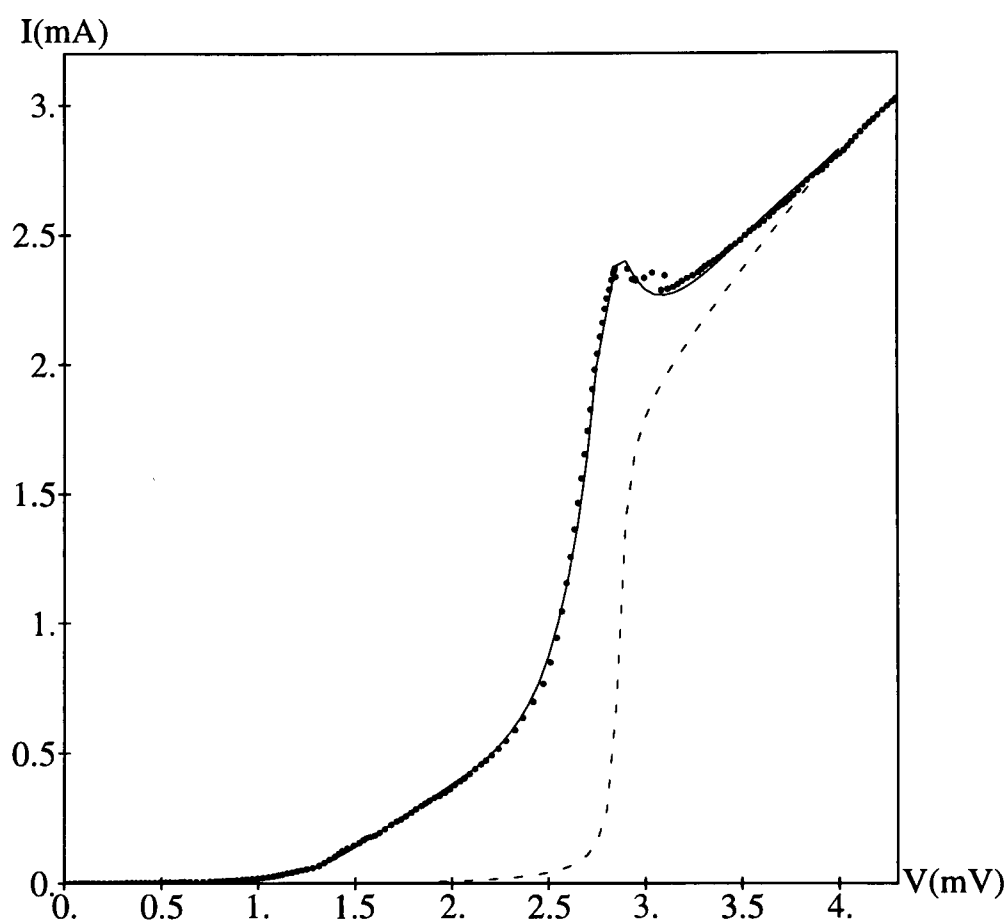


Figure 6.2.1 IV characteristic for a single crystal Nb–Ox–Pb tunnel junction (dots) at 1.6 K taken using a sweeping voltage bias circuit to highlight negative resistance region around 3 mV. The solid line is a fit to the IV generated by an empirical density of states illustrated in the next figure. The dashed line is the ideal IV expected on the basis of BCS density of states for both the Nb and Pb.¹⁴⁵

Figure 6.2.1 shows the IV characteristic at 1.6 K for a Nb_{bulk}–Nb_{ion damage}–Ox–Pb junction made on a single crystal Nb surface using Ar ion mill cleaning, prior to junction

¹⁴⁴The implantation depth of ions (1) in a target material (2) is given by

$$R_p(\text{\AA}) = \frac{60E(\text{keV})}{g_2(\text{gcm}^{-3})} \frac{(Z_1^2 + Z_2^2)^{1/2}}{Z_1 Z_2} \left(\frac{M_2(M_1 + M_2)}{M_1} \right) \left(1 + \frac{M_2}{3M_1} \right)^{-1}$$

where Z, M are the atomic numbers and relative atomic masses, g is the density and E is the energy of the impinging ions.

¹⁴⁵File: “DOS by Hand Fit 8.1.93” for Fig. 6.2.1 and 6.2.2.

fabrication, at the minimum ion kinetic energy of 2 keV¹⁴⁶ available in our evaporation system. Initial attempts were made to fit the IV using the McMillan proximity model. The fits were only in qualitative agreement, however, even with an introduction of an additional component of Nb_{normal}-I-Pb tunneling in parallel to the Nb_{bulk}-Nb_{proximity}-I-Pb process. As an alternative method we derived the Nb surface density of states, shown in Fig. 6.2.2, by guessing a form for the density of states, at 0.05 meV intervals, and comparing the resulting IV with the experimental one. The IV was calculated using Eq. 6.1.1 with $T = 1.6$ K and the Pb density of states given by the BCS form with a low temperature energy gap determined from previous experiments of $\Delta_{\text{Pb}}(0)^2 = (1.35 \text{ meV})^2 + 0.05i \text{ meV}^2$. This is shown as a solid line in Fig. 6.2.1.

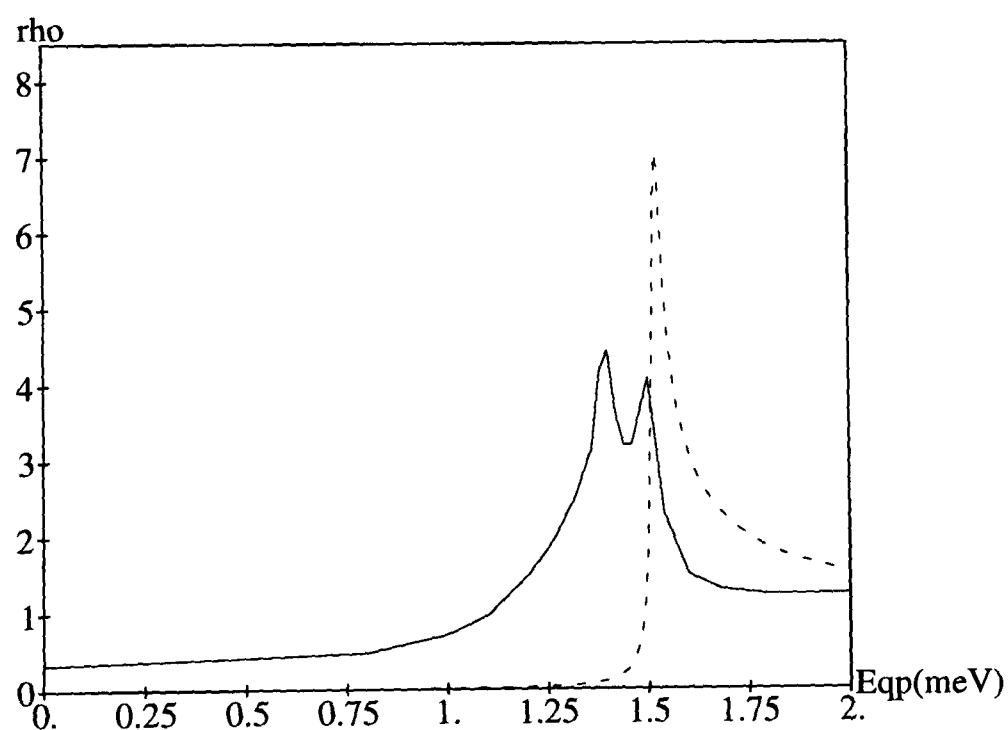


Figure 6.2.2 Empirically determined average density of states for the surface of the Nb crystal, ρ_S/ρ_N ¹⁴⁷, following ion milling by 2 keV Ar atoms and thermal oxidation. The resulting IV for Nb-Ox-Pb junction is shown in the previous figure. The theoretical BCS density of states for Nb ($\Delta = 1.515$ meV) is also shown.

¹⁴⁶Ion Tech Fast Atom Source: Kinetic Energy of Ar atoms is ~85% of operating voltage.

¹⁴⁷ ρ_N is the normal state density of states, ρ_S is the apparent superconducting density of states of the Nb crystal.

6.2.2. Comparison of Niobium Empirical Density of States with McMillan Proximity Model

If we now compare the double peak feature around 1.4 meV in the empirical density of states with the solutions from McMillan's Eqs. 6.1.10, 6.1.11 and 6.1.14 we can estimate a value for Γ_N , the scattering rate of quasiparticles from the bulk Nb into the damage layer. In making these calculations it was assumed that $\Delta_N^{ph} \equiv 0$, implying that the damage layer is a normal metal ($\lambda_N = 0$) and that $\Delta_S^{ph} = \Delta_{Nb}(T)$ ($\Delta_{Nb}(0) = 1.55$ meV). In practise, for the range of $\Gamma_{N,S}$ in this work the second approximation is reasonable and was confirmed by the non-iterative evaluation of the integral in Eq. 6.1.13. Since $N_S(0) = N_N(0)$ then Eqs. 6.1.8 and 6.1.9 gives

$$\frac{\Gamma_N}{\Gamma_S} = \frac{d_S}{d_N}. \quad [6.2.1]$$

This ratio is ~ 25 , if we assume the depth of the damage layer, d_N , created by the ion milling is that predicted kinematically, 13 Å, and that an appropriate value for d_S is the bulk Nb coherence length (~ 350 Å). The latter assumption is in keeping with the McMillan model since inserting the crystal dimension is clearly wrong. Figures 6.2.3 and 6.2.4 show the McMillan density of states and resulting IV for the closest match to the empirical density of states. The density of states was calculated with $\Gamma_N \approx 6$ meV and the other parameters are given in the figure caption.

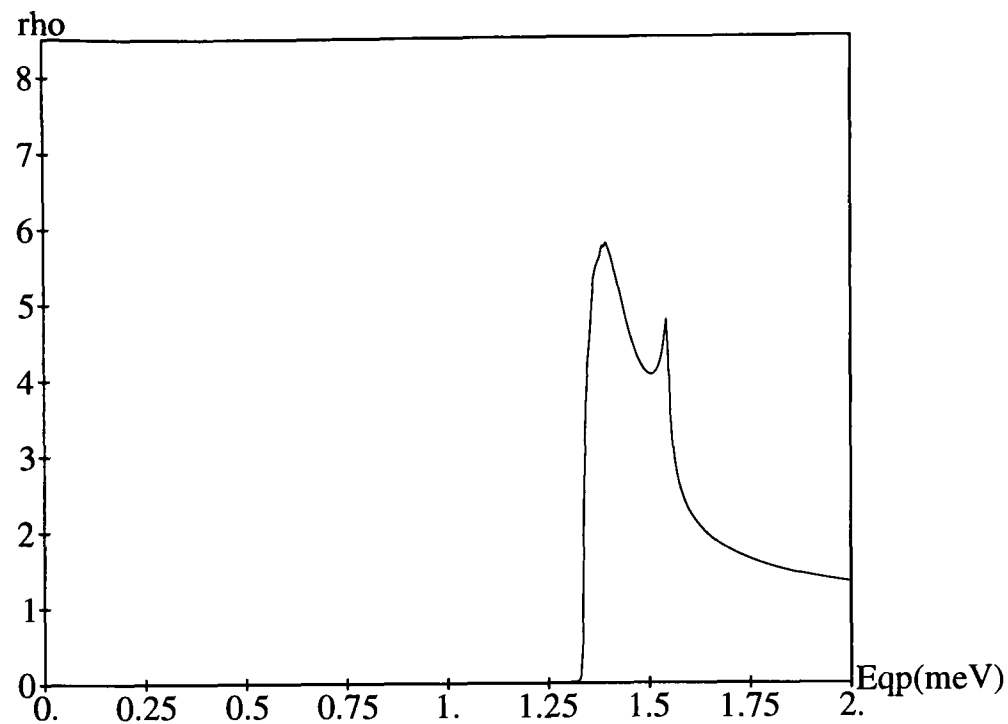


Figure 6.2.3 McMillan model density of states, ρ_S/ρ_N , for the best fit to the experimental Nb–Ox–Pb IV. The parameters used were $\Delta_N^{ph} = 0$, $\Delta_S^{ph} = 1.55$ meV, $\Gamma_N = 6$ meV, $\Gamma_S = 0.06$ meV.¹⁴⁸

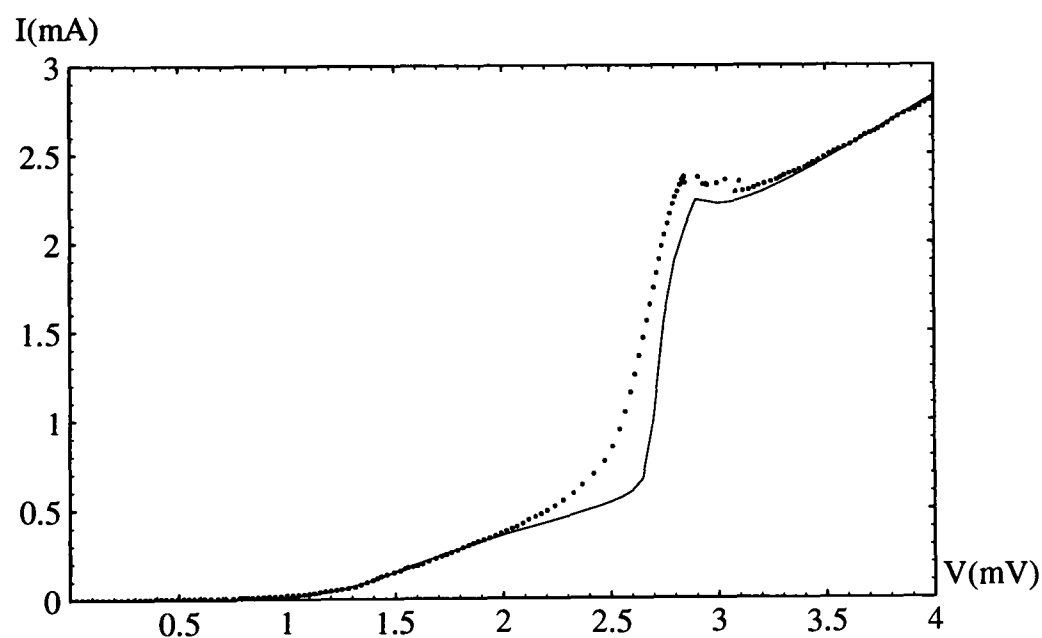


Figure 6.2.4 The IV characteristic generated from the McMillan density of states shown in the previous figure for a Nb–Ox–Pb junction (solid line). The experimental curve is shown as a dotted line. The parameters of the model were adjusted to give the best qualitative fit to the knee structure around 2.9 meV. The additional leakage current in the range 1–2 meV was fit by a 33% normal contribution in the Nb density of states (see discussion in §6.2.3). The Pb film is assumed to have a BCS density of states with $\Delta_{Pb}(0)^2 = (1.35 \text{ meV})^2 + 0.05i \text{ meV}^2$.

Using Eq. 6.1.8, with $\Gamma_N = 6$ meV, the transmission probability, σ , at the interface of the bulk Nb and damage region is estimated to be 0.2, assuming v_F , $5 \times 10^5 \text{ ms}^{-1}$ and $B = 2$. This is similar to that calculated by Keith and Leslie for a similar experimental arrangement.

¹⁴⁸File: “nb070292 Fit IV MTM 18.1.93” for this and following Figs. 6.2.3–6.2.7

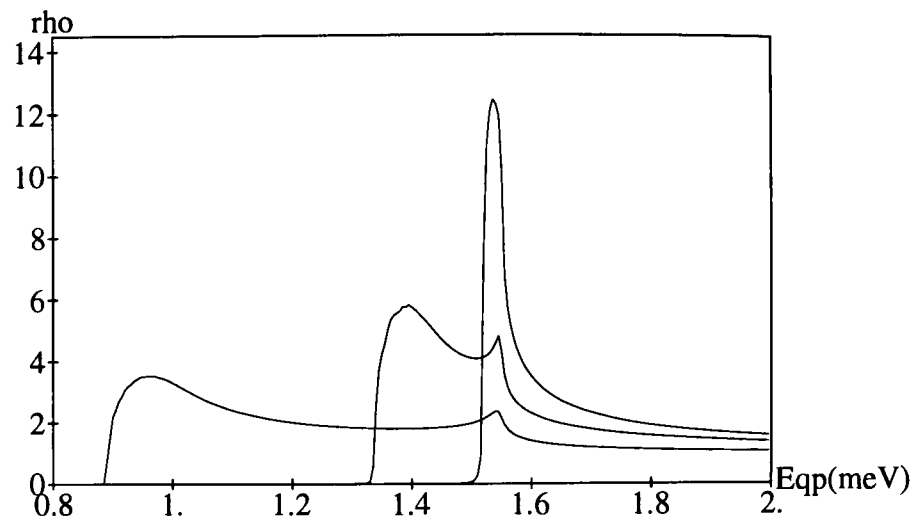


Figure 6.2.5 McMillan density of states, ρ_S/ρ_N , in the normal layer using parameters $\Delta_N^{ph} = 0$, $\Delta_S^{ph} = 1.55$ meV, $\Gamma_S = 0.06$ meV, $\Gamma_N = 2, 6$ and 20 meV (curves running left to right).

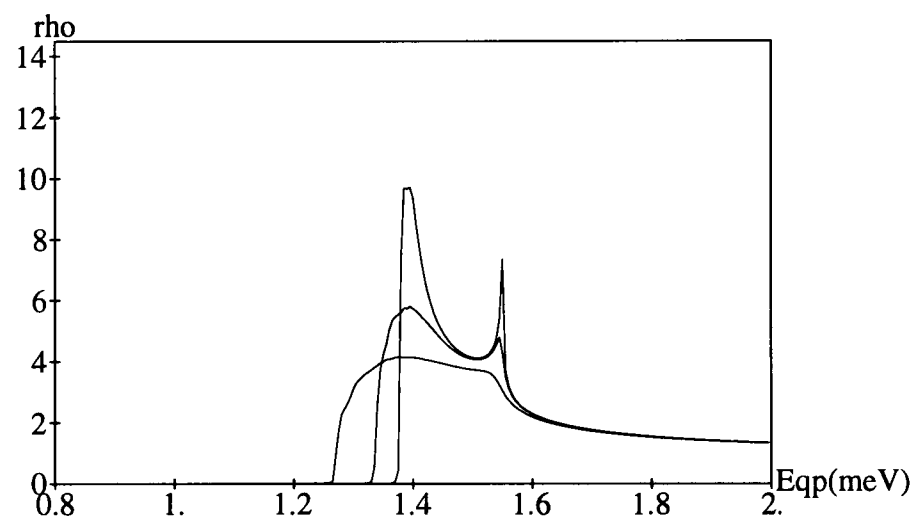


Figure 6.2.6 McMillan density of states, ρ_S/ρ_N , in normal layer using parameters $\Delta_N^{ph} = 0$, $\Delta_S^{ph} = 1.55$ meV, $\Gamma_N = 6$ meV, $\Gamma_S = 0.2, 0.06$ and 0.01 meV (curves running left to right).

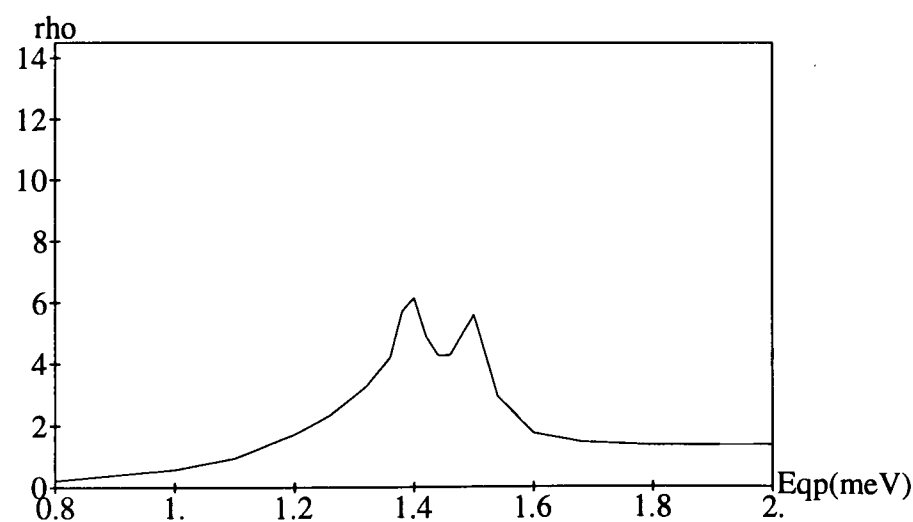


Figure 6.2.7 Empirical density of states, ρ_S/ρ_N , obtained from IV shown in Fig. 6.2.1. We have excluded a 33% normal component, and renormalised the result, to allow direct comparisons with figures above. Despite the subtraction of the normal component a tail at energies 0.8 – 1.3 meV is still present.

In light of the poor fit of the McMillan theory in regions away from the double peak structure it is instructive to look at the variation in the density of states in the normal

metal with Γ_N and Γ_S (see Figs. 6.2.5–6.2.7). As stated earlier the low energy peak is a very strong function of Γ_N and so fitting the shoulder structure of the IV around 2.9 meV (which is dependent on the peak separation) gives this value to relatively high accuracy. An attempt to vary Γ_S to improve the fit below the shoulder causes the shoulder itself to become washed out due to the lower of the two peaks becoming less well defined.

6.2.3. Explanation of Additional Structure in Niobium Surface Density of States

It remains for us to suggest explanations for two additional features shown by the Nb surface density of states (in Fig. 6.2.2) which was derived from the Nb–Ox–Pb STJ IV characteristic (shown in Fig. 6.2.1). Firstly, the apparent normal region at quasiparticle energies significantly below Δ_{Nb} causing the leakage current in the Nb–Ox–Pb STJ at and above Δ_{Pb} on the IV curve of Fig. 6.2.1. Secondly, the relatively slow fall in the density of states from the lower of the two peaks toward lower energies shown in Fig. 6.2.2. This second feature is in contrast to the sharp cut-off on the low energy side of the density of states predicted by BCS theory (see dotted curve in Fig. 6.2.2), behaviour which is also evident in McMillan solutions which generate a finite energy gap (see Fig. 6.2.5 and 6.2.6).

Low bias leakage currents have been observed previously in Nb–Ox–Pb and Nb–Ox–Nb tunnel junctions involving single crystal, polycrystalline and sputtered films. The normal tunneling fraction for the junction shown in Fig. 6.2.1 is 33% compared to values from the work of Keith and Leslie (1978), which show 25% for Nb_{foil}–Ox–Pb; Broom (1978), 33% for Nb_{sputtered film}–Ox–Pb; Frommer et al. (1974), ~30% for Nb_{single crystal}–Ox–Pb; MacVicar and Rose (1968), ~30% for Nb_{single crystal}–Ox–Pb; and Sherill and Edwards (1961), ~25% (with some samples very much higher) for Nb_{polycrystalline foil}–Ox–Pb. The magnitude of this leakage current is considerably higher than that which might arise due to multi-particle tunneling [136], even in high conductance devices. The measured fraction of leakage current cannot be used to estimate the percentage of the Nb

surface that is normal since the conductivity of such regions will be different from that of the conventional tunneling route for quasiparticles.

Halbritter [137] has proposed a surface mechanism for creating the normal tunnel regions, at the surface of a Nb crystal, which is due to the properties of the native oxide. At the metal–oxide (Nb_2O_5) interface a clear cut spatial separation in metallicly conducting regions and dielectric regions is impossible because of continuous stoichiometric changes and leakage of conducting electrons out of the metal, Φ_C , (over characteristic distances $\sim 2\text{--}20 \text{ \AA}$). Localised states, Φ_L , outside the metal hybridise with these leaking conducting electrons. Such surface states, where an extended Φ_C and a localised Φ_L component are mixed yield various surface effects including a weakening of the superconducting interaction. This creates both a depression in the observed superconducting energy gap and also normal regions due to a seam of such surface states. For Nb– Nb_2O_5 –metal junctions he estimates that the rate of exchange of electrons between Φ_C and Φ_L will be small compared to the inverse surface state lifetime with respect to tunneling to the counter electrode. Thus the surface states will act as holes in the tunneling barrier. He estimates the diameter of the holes is $\sim 5 \text{ \AA}$. The resulting gap depression will be $\sim 2\%$ and there will be a normal component in the density of states $\sim 15\%$. He is at pains to point out that the normal surface states do not interfere with the superconducting wave function. Thus he explains the commonly occurring IV characteristic in terms of Nb– Nb_2O_5 regions without taking the $\text{NbO}_{0.02}$ inclusions [138] and $5 \text{ \AA}$ thick NbO ¹⁴⁹ layers [139] into account. The last two oxides have been suggested as the cause of these effects by other authors.

In analysing sputtered Nb–Ox–Nb junction IVs Broom [130] proposed an empirical form for the additional density of states at the Nb surface¹⁵⁰. We find that this form with only minor modification of the free parameters is also able to make a good fit of our data

¹⁴⁹NbO in crystalline form is a superconductor, $T_c \sim 1 \text{ K}$. However, Halbritter points out that, in the amorphous form that it is probably present at on the surface of Nb, it will not superconduct.

¹⁵⁰Excess density of states given by,

$$\rho_{\text{excess}}(E) = K_1 \frac{|E|}{[E^2 - \Delta^2]^{\frac{1}{2}}} + K_2 e^{-(E/E_0)^2} \quad K_1 = 0.45, K_2 = 0.33, E_0 = 0.6 \text{ meV}$$

below the double peak structure. Since Broom offers no explanation for the form of this additional density of states it sheds no light on the cause of the structure except in so much as these features appear to be common between sputtered Nb films and Nb single crystals. Broom et al. [129] point out, in agreement with the conclusions of Keith and Leslie, that they can reduce the size of the shoulder structure they observe in sputtered Nb–Ox–Pb junctions by reducing the RF voltages used in sputter cleaning prior to their oxidation step.

Arnold [140] has considered proximity models (involving specular reflection at the boundary surfaces) for a thin normal metal backed by a semi infinite superconductor. In the limit that $d_N \ll \xi_0$ he predicts the existence of a smeared resonance state in the normal layer just below Δ_S and the form of his density of states appears to have some qualitative resemblance to our empirical one below the peaks toward lower energies. However, given the evident mix of effects that are present at the Nb surface it is difficult to confirm this interpretation of our results.

It is worth noting that the shoulder feature has been observed in the IVs of junctions examined in a number of different experiments. Keith and Leslie showed that the effect can arise from ion milling damage (and were able to significantly reduce the effect by using heavier bombarding atoms, reducing the damage layer to $\sim 1 \text{ \AA}$). Frommer et al. [141] saw similar shoulder structures in junctions on Nb single crystals where no ion milling/sputter cleaning was performed. Their crystals were grown in ultra-high vacuum by the floating zone electron beam method. The crystal was then allowed to cool in situ and oxygen introduced directly in to apparatus to create the thermal oxide barrier. The cooling period itself was found to be the major determinant of final junction quality. If cooling extend past the mono-layer time (of the back ground atmosphere ~ 6 hours) a deterioration in barrier performance was observed. However, even in the rapidly cooled devices features at the shoulder of the IV were observed and were thought to be caused by “adsorbed oxygen”. In Pb–Ox–Pb junctions Wyatt et al. [142] were able to reproduce the shoulder feature by adding a mono-layer of Zn between the first Pb and the oxide barrier.

They were able to successfully analyse their IVs in terms of the McMillan proximity model. These junctions showed no sign of any sub gap leakage current.

In summary, the shoulder structure and subgap leakage currents of STJs fabricated from Nb appear to be intrinsic properties of the Nb. While, some progress has been made in reducing these features [122], the thin film techniques involved are not suitable to our experiments with single crystal Nb. Our experiments will have to be conducted in the light of the disappointing conclusion that the “clean” Nb surface represents a far from ideal regime in which to study the superconductivity of bulk single crystal Nb.

6.3. SUPERCONDUCTING PROXIMITY LAYERS ON NIOBIUM SINGLE CRYSTAL SURFACES

6.3.1. Introduction

We have investigated the behaviour of thin (50–2000 Å) Al layers on the surface of single crystal Nb samples. These geometries were designed to investigate quasiparticle trapping from the Nb crystal into the Al thin film. As reported in §5 these investigations were inconclusive. Those Al traps fabricated in our evaporation system directly prior to junction fabrication showed extremely poor quality interfaces between the Al and Nb crystal. The reason for the failure to observe quasiparticle trapping in these devices is probably related to the poor transmission coefficient between the thin film and crystal. A significant improvement in the quality of the Nb–Al interface was achieved in a separate CVC sputtering system with a base pressure ($\sim 2 \times 10^{-8}$ torr) two orders of magnitude better than the evaporator and importantly an oxygen partial pressure $\sim 10^3$ lower.

In this section we will discuss the experimentally observed Al proximity layer density of states and attempt to apply the McMillan tunneling model once again.

The analysis of Al proximity layers is similar to that outlined in the previous section for the Nb damage layer. However, in looking to derive solutions for the Al proximity layer density of states we must include an estimate of the electron–phonon coupling strength in the Al which enters into the self-consistent condition, Eq. 6.1.12. In the case

of thick films ($\geq 1000 \text{ \AA}$) we will assume the values, $\lambda = 0.171$ and $\omega_N = 29.95 \text{ meV}$, which are consistent with the bulk energy gap of $170 \text{ \mu eV}$. For Al films thinner than this the energy gap becomes elevated. This has been observed experimentally for both very thin films and also granular Al films. In order to fit our experimental data in this case we will allow the energy gap (in the absence of the proximity effect) as a floating parameter.

6.3.2. Aluminium Proximity Films Fabricated in the Evaporation System: Poor Nb–Al Interfaces

The McMillan model predicts that for a finite transmission coefficient at the Nb–Al interface a gap will exist in the Al proximity layer at temperatures both below and above the Al thin film T_c . This will be apparent in the IVs of junctions made on the proximity films. Figure 6.3.1 shows the temperature variation of the gap measured using a Pb probe film on four separate Al proximity layers fabricated entirely in our evaporation system. In addition to the low energy gap McMillan's model predicts there will also be a small anomaly in the proximity film density of states around Δ_{Nb} . However, this feature is too small to observe in the temperature range considered.

For Al films of thickness $\sim 60 \text{ \AA}$ the ratio of Γ_N to Γ_S is given by Eqs. 6.1.2 and 6.1.3

$$\frac{\Gamma_N}{\Gamma_S} = \frac{d_S N_S(0)}{d_N N_N(0)} \approx 15, \quad [6.3.1]$$

where $N_S(0)$ and $N_N(0)$ are the bulk values for Nb and Al of 31.7 and 12.2×10^{21} states $\text{eV}^{-1} \text{ cm}^{-3}$ and, again, we assume $d_S \sim \xi_0$. The latter assumption is not critical since the low energy gap values derived from the McMillan model are insensitive to the precise value of Γ_S below values of $\sim 0.1 \text{ meV}$.

Attempts to fit the observed temperature dependence of the induced Al energy gap were only partially successful. If the bulk value of the Al electron–phonon interaction was assumed, agreement with the low temperature proximity energy gap was achieved with $\Gamma_N = 0.06 \text{ meV}$, but with the theoretical curve showing a much shallower slope than that exhibited by the experimental data in the range $2\text{--}3 \text{ K}$. Some improvement in the quality

of fit was achieved by assuming the Al energy gap was somewhat enhanced, over the bulk value, to 250 μeV . Figure 6.3.1 shows the energy gap temperature dependence, assuming a 250 μeV Al energy gap, for $\Gamma_N = 0.025$ and 0.04 meV. The other parameters were $\Gamma_S = 0.001$ meV, $\omega_S = 23.9$ meV, $\lambda_S = 0.2918$, $\omega_N = 43.32$ meV, $\lambda_S = 0.171$ ¹⁵¹. The agreement is improved but still leads us to conclude that the McMillan model is unable to adequately explain the observed temperature dependence for these devices. If we assume that the McMillan solutions give some indication of the quality of the Nb–Al interface then $\Gamma_N = 0.04$ meV corresponds to a transmission coefficient of 6×10^{-3} .

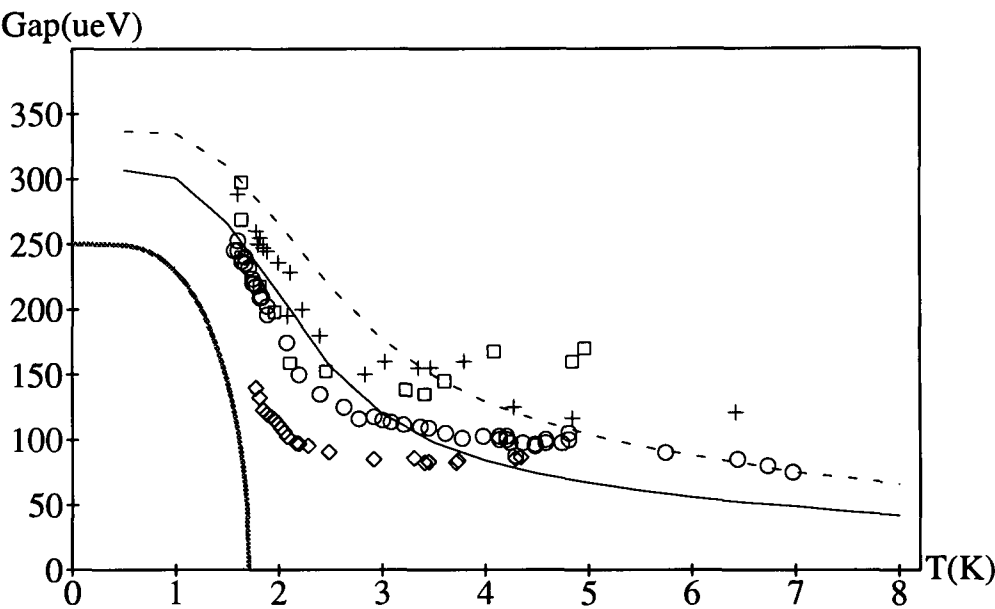


Figure 6.3.1 The temperature dependence of the energy gap induced in a thin film of Al on the surface of a single crystal of Nb. The Al films had thicknesses and deposition rates of: Circles 60 Å @ 30 Å s⁻¹; Diamonds 60 Å @ 25 Å s⁻¹; Crosses 60 Å @ 20 Å s⁻¹; Squares 80 Å @ 12 Å s⁻¹. The lines are theoretical temperature dependencies from the McMillan model: Solid line $\Gamma_N = 0.025$ meV; Dashed line $\Gamma_N = 0.04$ meV; $\Gamma_N/\Gamma_S = 15$. Other parameters are given in the text. The grey line in the left hand corner is the BCS temperature dependence for an Al thin film with $\Delta_{\text{Al}} = 250 \mu\text{eV}$.¹⁵²

Surprisingly, for a given Al thickness slightly better Nb–Al transmission coefficients were obtained for lower Al deposition rates. These devices were fabricated in a thin film evaporation system with a base pressure of $\sim 10^{-6}$ torr (see §4). At this pressure the mono-layer time is less than 1 second. Despite outgassing of the Al source boats prior to use this pressure was still observed to rise by ~ 10 during the trap deposition. It seems

¹⁵¹In the limit that Γ_N becomes very small, corresponding to the transmission coefficient between the Nb–Al layer ≈ 0 , the expected temperature dependence below the T_c of Al will be close to that of BCS predictions.[135] This is shown by the grey line in the bottom left-hand corner of Fig. 6.9.
¹⁵²File: “Induced Proximity Gap 26.1.93”

likely that during the delay between ion milling the Nb surface and trap deposition (~1 minute) there is considerable contamination of the interface which was made worse by the heating of the Al boat. In the identical experimental set up for In single crystals Goldie [31] typically observed transmission coefficients for In single crystal–Al proximity layers ~0.2. As has been mentioned previously the short coherence length of Nb makes this material much more sensitive to interface contamination. The coherence length of In is ~10 times that of Nb and so is more tolerant of interface contamination. Additionally, the formation of In oxides¹⁵³ takes place much more slowly than those for Nb. In order to reduce these effects we investigate the fabrication of Al proximity layers on Nb crystals in a better vacuum system.

6.3.3. Aluminium Proximity Films Fabricated in the CVC Sputtering System: Good Nb–Al Interfaces

We were able to significantly improve the Nb single crystal–Al interface by using a CVC sputtering system located in the Oxford Cryodetector Group clean room. The CVC sputtering system is normally used for fabrication of Nb and Al thin film devices, patterned using photolithographic techniques. This system is able to achieve oxygen partial pressures that are considerably better than those available in our evaporation system (see §4). Following the deposition of an Al proximity layer the Nb crystals still had to be transferred to the evaporator, for the fabrication of junctions, exposing them to atmosphere. In initial tests this lead to single junction normal state resistances greater than 10 k Ω . While we were able to make tests of the IV characteristics of these devices the high dynamic resistance made performing measurements of non–thermal signal propagation in the Nb crystals difficult. This method of fabrication remains extremely encouraging for the observation of quasiparticle trapping from Nb single crystal devices but no results, as yet, can be reported.

¹⁵³In general the oxygen partial pressure in the evaporation system was ~10% of the base pressure just prior to Al layer deposition.

In initial tests in this system we deposited Al thin films of $\sim 80 \text{ \AA}$ for direct comparison with those reported in the previous section. Figure 6.3.2 shows a typical IV for such a Nb–Al–Ox–Pb device. The apparent gap of the Al proximity film was close to that of the bulk Nb indicating a significantly improved transmission probability at the Nb–Al interface. This can be seen by first comparing the IV with that for an evaporator fabricated Al proximity layer (short dashed line in Fig. 6.3.2) and that for a Pb probe film directly on the surface of the crystal (dashed line). Fits based on McMillan model generated IVs yield a value of Γ_N consistent with $\sigma \sim 0.2\text{--}1$ where the uncertainty arises from the errors in the determination of the Pb energy gap.

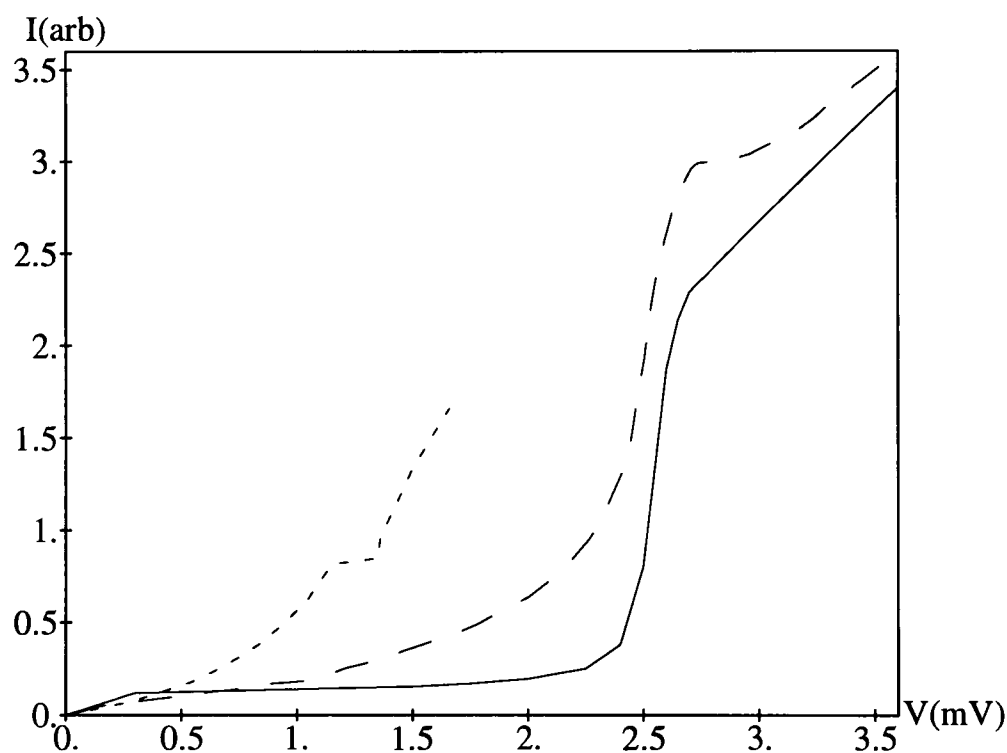


Figure 6.3.2 Comparison of current–voltage characteristics of Nb–Ox–Pb (dashed line) at 4.2 K, Nb–Al(60 $\text{\AA}$)–Ox–Pb (short dashed line) at 4.2 K with trap made in the evaporation system and Nb–Al(80 $\text{\AA}$)–Ox–Pb (line) at 4.8 K with trap made in CVC sputtering system. The IVs have been normalised to have equal normal state resistances. The actual values were 1.4 Ω , 2.3 k Ω and 80 k Ω .¹⁵⁴

Figure 6.3.3 shows the temperature dependence of the energy gap induced in a 2000 $\text{\AA}$ Al film deposited in the CVC system. In this case a fit using the predicted gap variation of the McMillan model gives extremely close agreement. The best fit was achieved for $\Gamma_N = 0.06 \pm 0.01$. The other parameters were $\Gamma_S \leq 2 \Gamma_N$ ¹⁵⁵, $\omega_S = 23.9 \text{ meV}$,

¹⁵⁴File: “STJ IV Comparison 31.1.93”

¹⁵⁵The fit is insensitive to this parameter in this range.

$\lambda_S = 0.2918$, $\omega_N = 29.95$ meV, $\lambda_S = 0.171$. The last two values being consistent with the bulk Al energy gap of $170 \mu\text{eV}$. This value of Γ_N , using Eq. 6.1.8, gives a transmission coefficient at the Nb–Al interface of 0.29 ± 0.05 .

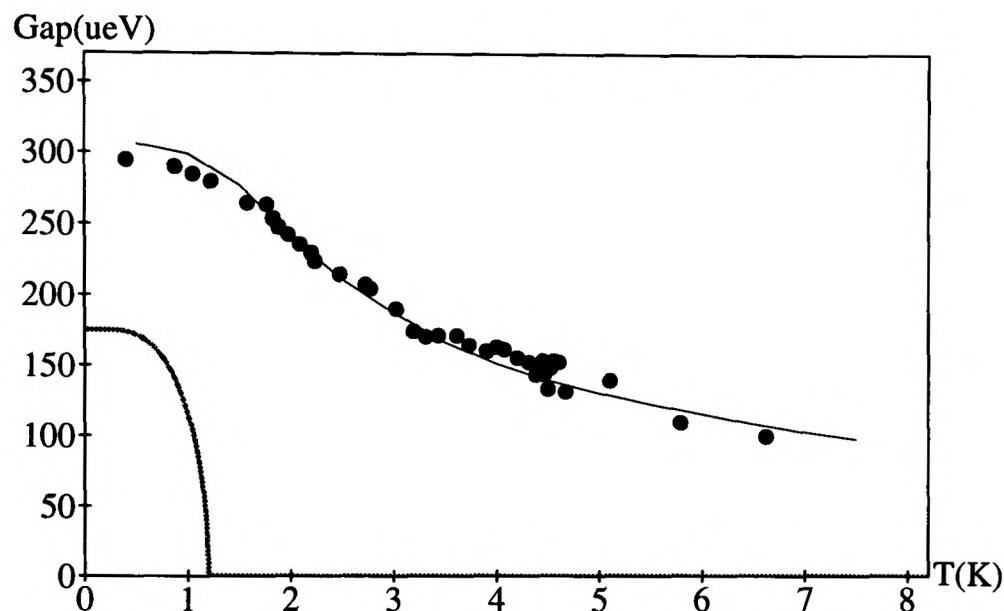


Figure 6.3.3 Temperature variation in energy gap induced in $2000 \text{ \AA}$ Al proximity film, fabricated in CVC sputtering system. Line is fit using predictions of McMillan model for $\Gamma_N = 0.06$ meV with other parameters given in the text. The curve in the bottom left-hand corner is the BCS temperature dependence of the low energy gap for the Al in the absence of the proximity effect.¹⁵⁶

6.4. CONCLUSION

Nb–Ox–Pb junctions made directly on the surface of the Nb crystals showed significant leakage currents and shoulder characteristics at the sum of the energy gaps that indicated surface damage due to milling and also non-ideal tunneling barriers formed from the native oxides.

Initial attempts at fabricating Al proximity layers, in our evaporation system, on Nb single crystals were frustrated by surface contamination of the interfaces between the Nb and Al in a vacuum system with a base pressure $\sim 10^{-6}$ torr.

Progress was made in improving the quality of the Nb–Al interfaces by proximity layer fabrication in a system with much higher quality vacuum than our original evaporation system. By using a combination of low energy Ar ion milling (0.5 keV) and

¹⁵⁶File: "Induced Proximity Gap 26.1.93"

Al deposition in the CVC sputtering system we were able to create proximity layers with transmission coefficients ~ 0.3 . In the case of thin Al (80 Å) films the IV characteristics showed a proximity layer density of states close to that of bulk Nb without the leakage currents associated with native oxides. This technique is now common place in the fabrication of Nb thin film superconducting devices where Nb–Al–Ox–Al–Nb “tri-layers” are used combining the benefits of the high T_c of Nb with the robustness of AlOx tunneling barriers. At present the normal state resistances of our Nb–Al–Ox–Pb junctions are very high ($\gg 10\text{ k}\Omega$). We would hope to reduce these values in future work.

These results are encouraging for the application of quasiparticle trapping from bulk Nb single crystals. The finite ($\sim 10\text{ }\mu\text{s}$) propagation times of quasiparticles over distance $\sim\text{cm}$, in the extremely high purity Nb samples used in these experiments, will still limit the effective rate at which quasiparticles can be trapped but we believe that we will soon be able to unambiguously differentiate between quasiparticle and the pervasive phonon signal arising in STJs and SASTJ. This is a crucial criterion if we are to use the ratio of these signals to gain further information on the nature of the original particle interaction in the Nb crystal and so possible differentiate between nuclear recoil and electron events.

7. PARTICLE DETECTION

It seems to me that what can happen in the future is ... that the experiments get harder and harder to make, more and more expensive ... and it [scientific discovery] gets slower and slower.

Richard Phillips Feynman

7.1. INTRODUCTION

In this chapter we discuss the detection of α -particles and γ -rays in single crystals of superconducting Nb of masses 2 and 12 g. The non-equilibrium phonon and quasiparticle states arising from particle interactions therein were observed at crystal surfaces using series arrays of Al superconducting tunnel junctions (Al SASTJs, see §4.3.3). The experiments were conducted in a ^3He cryostat (§4.4.1) at a temperature of 360 mK. 5.5 MeV α -particle interactions were observed in the 12 g (length 15 mm $\times$ diameter 10 mm) crystal with an energy resolution of 170 keV FWHM and a position resolution of better than 500 μm . We have also observed position dependent non-equilibrium signals arising from α -particles and γ -rays in 2 g crystals. Ultimately, we are interested in the possibility of differentiating between nuclear and electron recoil events in Nb if it is to be used as a dark matter detector (see Chapter 2). In light of the results presented here we discuss techniques that may allow us to differentiate between these types of interaction.

Early experiments on the (un-outgassed) low purity Nb crystals showed that the signal shapes detected for laser and α -particle signals were identical [119]. The experiment was performed on a Nb crystal (length 2 mm $\times$ diameter 10 mm) using an Al SASTJ fabricated on the opposite face to which was excited by the laser/ α -particle. Figure 7.1.1 shows the SASTJ voltage signals at 360 mK (averaged over 50 events, in each case, to improve the signal to noise ratio). The phonon and quasiparticle diffusion signals could be clearly identified due to the significant difference in their propagation times. The phonon signal precedes that of the quasiparticles ¹⁵⁷.

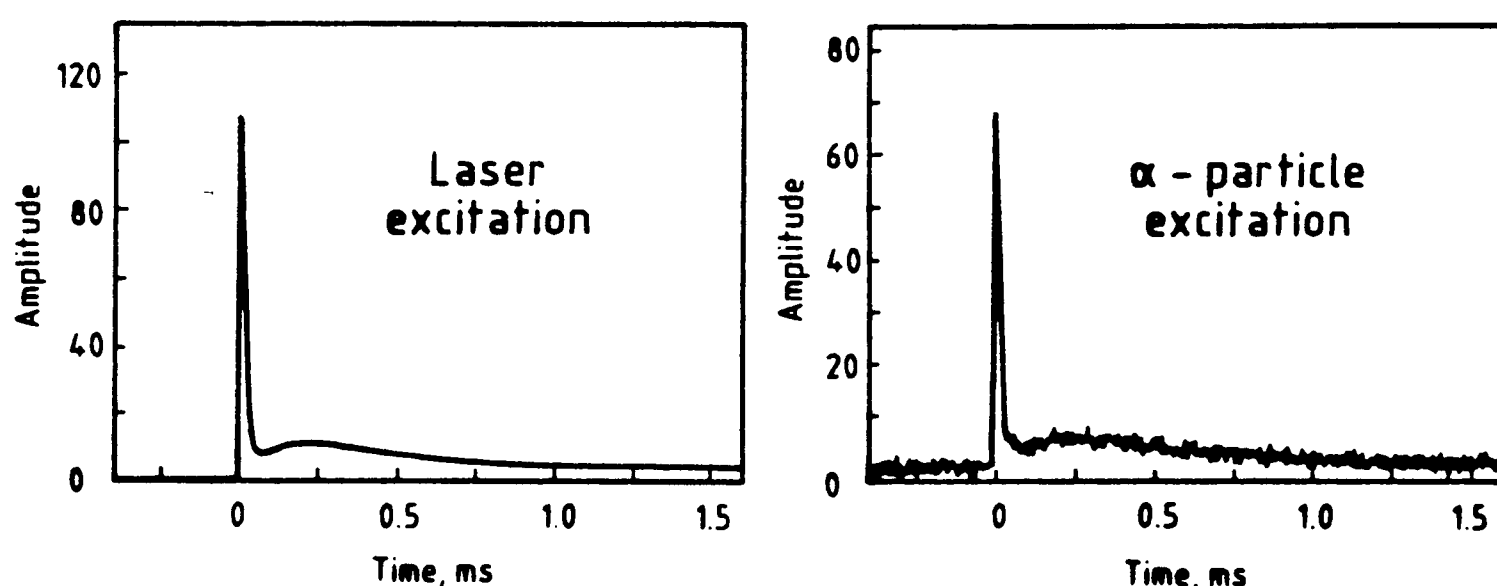


Figure 7.1.1 Comparison of the voltage pulses due to laser excitation and α -particle excitation of a low purity Nb crystal (length 2 mm $\times$ diameter 10 mm, not outgassed). The sensor was an Al SASTJ (200 junctions). The pulses decay at long time-scales due to an electronics ac coupling (CR) time of 900 μ s. The signals were averaged over 50 events to improve the signal to noise ratio.[119]

In the high purity Nb crystals the propagation times of the phonons and quasiparticles becomes comparable. It is no longer possible to measure the relative signal sizes using time separation techniques. We are investigating alternative techniques (discussed in §5.4) to clearly distinguish between the signals. For the results presented in

¹⁵⁷This is discussed in detail in Chapter 5.

this Chapter we believe that the geometries are sensitive to quasiparticle signals, however, we have yet to demonstrate this unambiguously.

7.2. DETECTION OF ALPHA PARTICLES IN A 12g NIOBIUM CRYSTAL

Two pairs of Al SASTJs were fabricated on the opposite faces of a 12 g cylinder of single crystal Nb, as indicated in Fig. 7.2.1. An Al quasiparticle trap, 500 Å thick, was deposited at one end of the crystal to try to improve the sensitivity of that pair of arrays to the quasiparticle signal arising from the interaction. The crystal was clamped in a copper block and cooled to a temperature of 360 mK. The lower edge of the crystal was then irradiated with 5.5 MeV α -particles collimated through 5 holes.

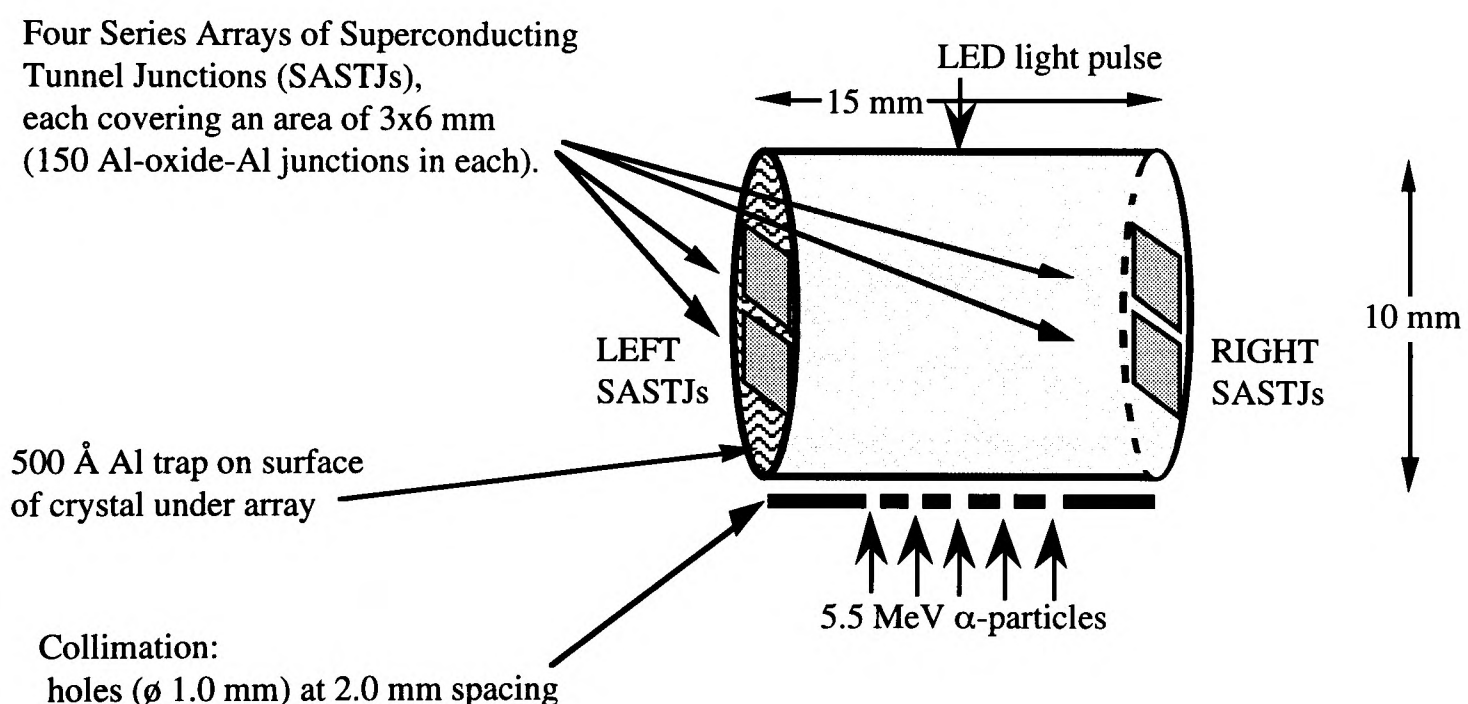


Figure 7.2.1 Irradiation of 12 g Nb crystal by collimated α -particle source. Events were recorded at 360 mK.

Four terminal connections from each array were brought to a voltage amplifier (see §4.5) outside the cryostat. In this experiment the two arrays, at each end of the crystal, were connected in series at the amplifier. The left and right composite arrays were then current biased. The voltage signals observed for an alpha interaction defined by the far

right hand hole of the collimator are illustrated in Fig. 7.2.2.¹⁵⁸ The signal from a SASTJ is a convolution of the phonon energy entering the SASTJ and breaking pairs in the Al thin films combined with the electronic RC time constant ($\tau_{RC} < 1 \mu s$) and the effective recombination time of the quasiparticles in the Al ($\tau_r \sim 20 \mu s$)¹⁵⁹.

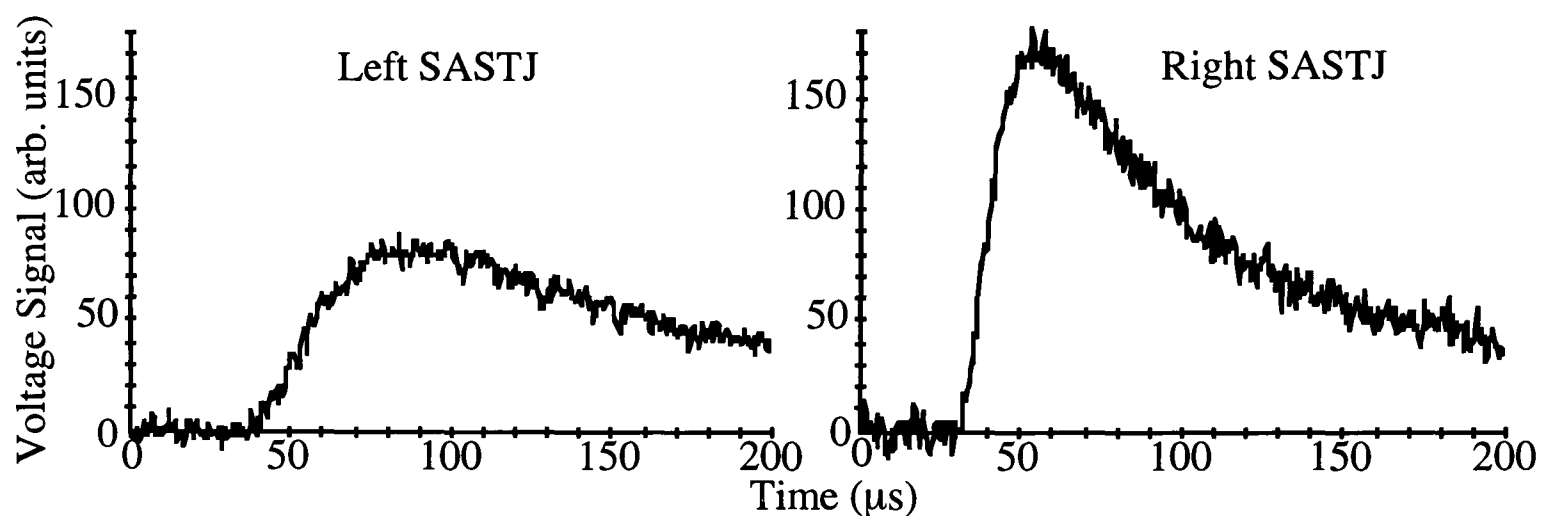


Figure 7.2.2 Voltage signals from constant current biased SASTJs at opposite ends of the crystal following an α interaction defined by the hole nearest the right hand end. The asymmetry in pulse height is due to the different propagation distances from the hole to the left and right arrays.

A scatter-plot of the voltage signal heights, in the left versus right SASTJs, arising from the interaction of α -particles at positions defined by the five hole collimator (1 mm diameter holes at 2 mm spacing) is shown in Fig. 7.2.3. The plot also includes events that were generated by periodic light pulses from a low power LED¹⁶⁰ which were directed by a quartz fibre on to the upper surface of the Nb crystal directly opposite the central hole as indicated in Fig. 7.2.1. These pulses permitted the monitoring of the stability of the array sensitivity and give an estimate of the intrinsic scatter, due to electronic noise, of the signal heights.

It is evident, from Fig. 7.2.3, that the peak voltage signal height in a SASTJ is a function of its distance from the interaction point. This would be expected since the signal

¹⁵⁸The voltage signals were very similar in shape to those seen for laser illumination through the same hole in a previous experiment under the same bias and temperature conditions.

¹⁵⁹The convolution of the detector response is discussed in §3.6.1.

¹⁶⁰This is a different light source to the much higher power laser used in §5.

shape in the first $\sim 60 \mu\text{s}$ ¹⁶¹ is determined by the direct ballistic phonon flux¹⁶² and also the signals due to the high energy phonons (and quasiparticles) which diffuse¹⁶³. Using the signal heights alone the detector is clearly able to resolve the events corresponding to interactions in the five separate holes which are spaced at 2 mm intervals.

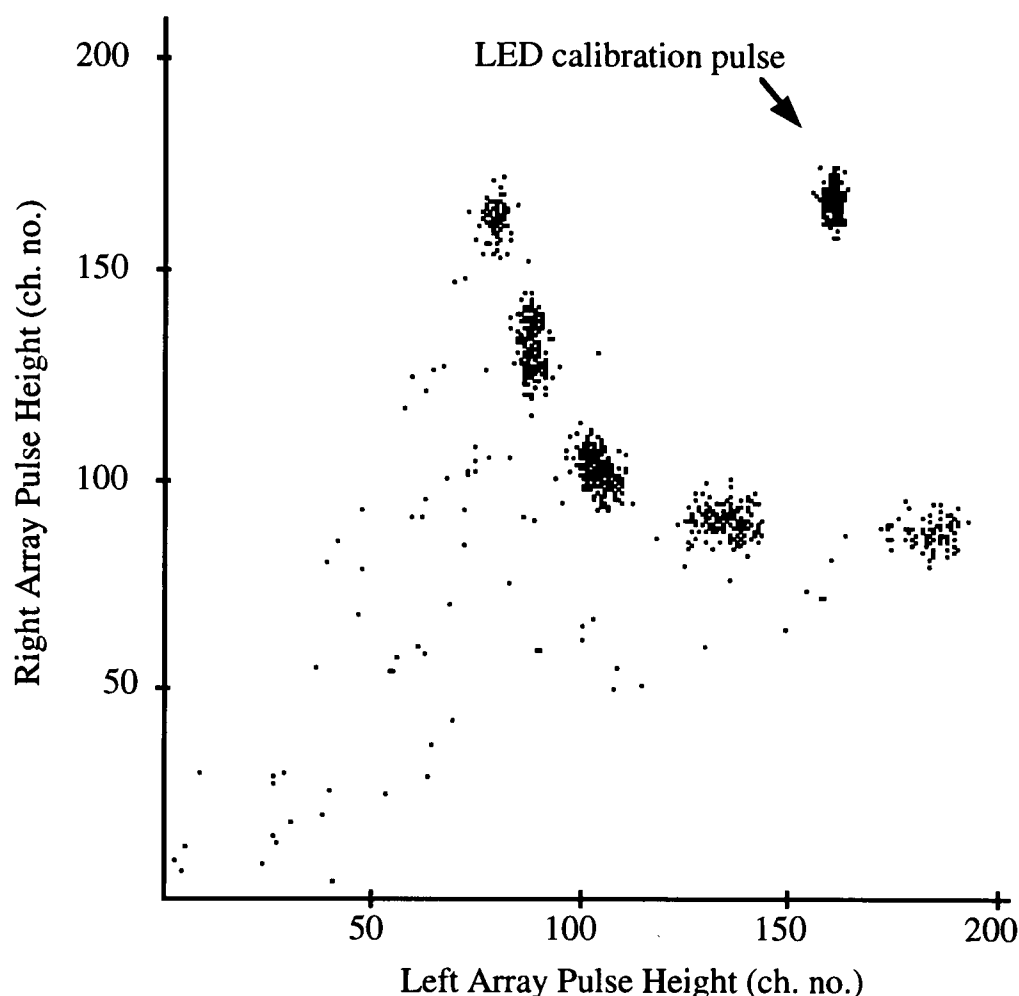


Figure 7.2.3 The pulse heights, in arrays at opposite ends of a 12 g crystal, due to 5.5 MeV α -particles (see Fig. 7.2.1) are plotted. The grouping in the top right hand corner is due to a LED calibration pulse incident on the crystal adjacent to the centre hole and gives an estimate of the signal/noise.¹⁶⁴

Closer analysis of the events from the centre hole shows that we can also see a residual inverse-correlation between the signal heights from the left and right SASTJs that must be due to the variation of the interaction position within the finite size of the collimator hole (diameter 1mm). Given the symmetry of the detector at this hole a crude

¹⁶¹It should be remembered that the integrating effect of the SASTJ response function (due to the finite recombination time (τ_r) of the Al quasiparticles) means that the phonon flux, at a given time, will influence the voltage pulse shape at later times (Δt), diminishing as $\exp(-\Delta t/\tau_r)$.

¹⁶²In the absence of attenuation, a ballistic signal would scale as (distance)⁻² due to the decrease in solid angle subtended by the detector.

¹⁶³In the absence of reflection from the surfaces of the crystal the peak diffusion signal height would show a (distance)⁻³ dependence. The pulse behaviour becomes more complex if reflection is assumed. This will be discussed in a later section.

¹⁶⁴File: "Alpha T001 (Heights)" nb020891.

ratio of the signal heights will give the position of the interaction independent of its energy. Comparison of the variation in this ratio for the α -particle events with that for the LED diode events (which acts effectively as a point source) imply an electronic noise limited position resolution of $\sim 300\text{ }\mu\text{m}$. If this position dependence is folded out of the pulse heights the resulting energy spectrum (for the central hole only), shown in Fig. 7.2.4, gives an energy resolution of 170 keV FWHM for the 5.5 MeV α -particles. Clearly, such an analysis is simplistic. A more complex functions (of the left and right signal heights) could be empirically determined for the position of the interaction by a fit to the centroids of the five separate point-distributions. The centroids lie on a mono-energetic locus at 2 mm intervals. We will discuss this in the later paragraph.

A slight asymmetry in the trend of signal heights with position for the left and right arrays was observed. This may be due to the enhanced sensitivity of the left hand array to quasiparticle trapping in the Al layer directly beneath it. However, the results detailed in Chapter 6 indicate that the technique used in the manufacture of this trap would be small due to a low transmission coefficient at the trap-crystal interface. Improvements have been made in trap fabrication (§6.3.3) and will be applied to future devices.

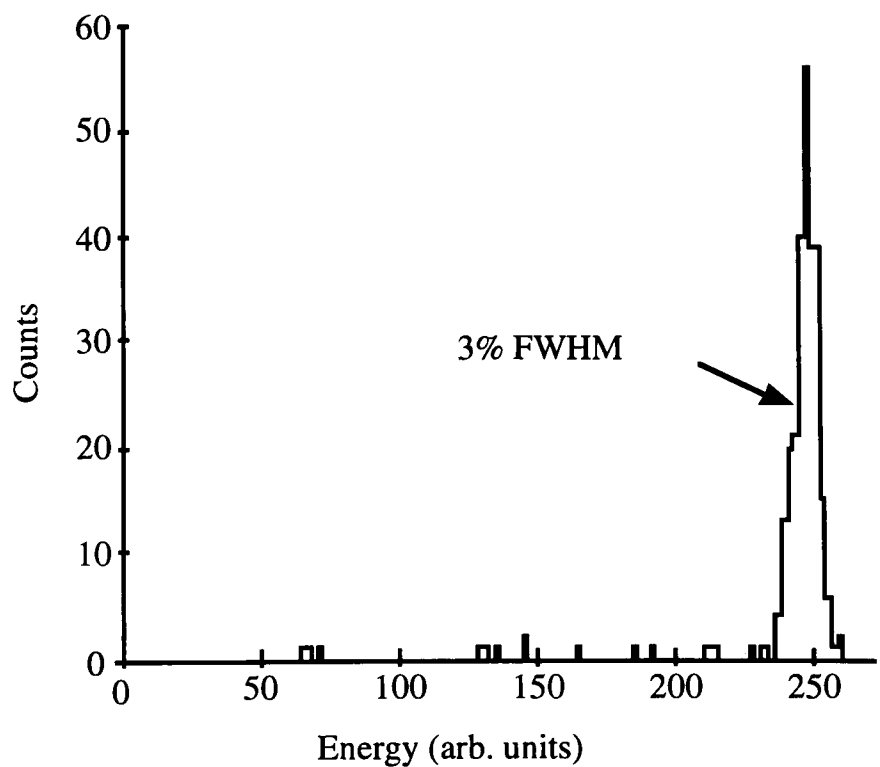


Figure 7.2.4 Plot of counts versus energy for 5.5 MeV α -particles from the centre hole giving an energy resolution of 170 keV FWHM.

The position of the interaction can also be determined by the difference in the arrival times of the signals in the left and right hand arrays. Figure 7.2.5 shows the timing difference extracted from the steepest parts of the leading edge of the voltage pulses ¹⁶⁵. The LED gives a FWHM position resolution of 300 μm . The resolution on the α -particle interactions appears to be $\sim 500 \mu\text{m}$.

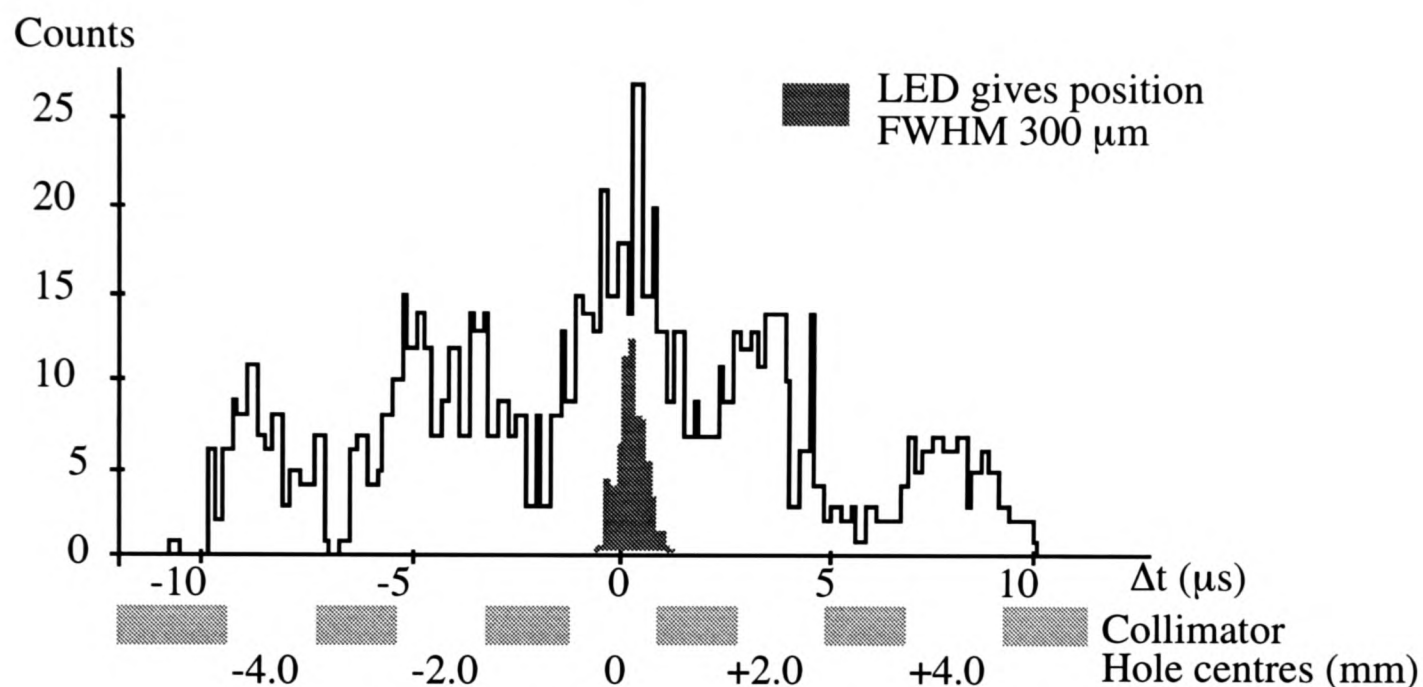


Figure 7.2.5 Direct measurement of difference in timing of steepest part of leading edge for signals in the left and right arrays.

The time difference of the earliest parts of the signals in the left and right arrays appears to be characteristic of slow transverse mode phonons (within the limits of the signal to noise). They appear to be produced in preference to longitudinal and fast transverse mode phonons for α -particle interactions. The overall voltage signal shapes (in the range 0–200 μs) and the variation of pulse height with distance could be fit using the diffusion simulation detailed in §3.5. The diffusion coefficient was fixed at $1.4 \text{ m}^2\text{s}^{-1}$ with an effective absorption at the crystal surfaces of 0.015. The use of a single diffusion coefficient to characterise the mixture of ballistic and range of diffuse components is an over-simplification, however, until we have a more sophisticated model it will have to suffice.

¹⁶⁵It should be reiterated that the voltage signal represents a convolution of the phonon flux and detector response.

The peak voltage signals recorded in a SASTJ, for the interaction of an α -particle at the furthest hole from it, corresponds to a phonon energy flux per junction of $8 \text{ eV } \mu\text{s}^{-1}$. We have used the same calculation as that reported in §5.5.7. If this flux figure is scaled by the ratio of the α -particle energy to that of the laser pulses (used in experiments reported in Chapter 5) it is shown to be in agreement to that recorded in §5.5.8 within a factor of 2.

It is apparent that the shape of the leading edge of the pulses is a function of the position of the interaction. If the energy spectrum of the phonons or the ratio of phonons to quasiparticles were to alter (due to a different type of interaction in the Nb) the position dependence of the pulse shape would alter. Such a change may provide a method to differentiate between a nuclear recoil and a γ -ray event. We intend to perform an experiment using a neutron source in the near future.

7.3. DETECTION OF ALPHA PARTICLE AND GAMMA RADIATION IN 2g NIOBIUM CRYSTALS

In order to investigate the behaviour of large mass Nb detectors to a range of particle radiations, a smaller crystal of mass 2 g (length 3 mm, diameter 10 mm) was used to improve the detector sensitivity at temperatures $\geq 360 \text{ mK}$. Al traps $1000 \text{ \AA}$ thick were deposited on both faces of the crystal and then SASTJs fabricated in the paired geometry similar to that for the 12 g device shown in Fig. 7.2.1.

Figure 7.3.1 shows the pulse heights from two SASTJs side-by-side on the opposite face to that which is exposed to 5.5 MeV α -particles collimated by a long slot. The pulse heights show a strong position dependence in a similar fashion to those for the 12 g crystal. The two apexes (marked as $\pm 1.5 \text{ mm}$) in the locus of constant energy occur when the α -particle interacts directly opposite the centre of one or other of the arrays. Outside this region both pulse heights decrease as the position of the interaction moves away from the array centres. As a consequence, the separation of the position and energy dependence of an event outside the central region is made difficult using the peak heights alone, but

this can be achieved with additional energy information as discussed later. The size of the position sensitive region can be increased by altering the geometry of the arrays.

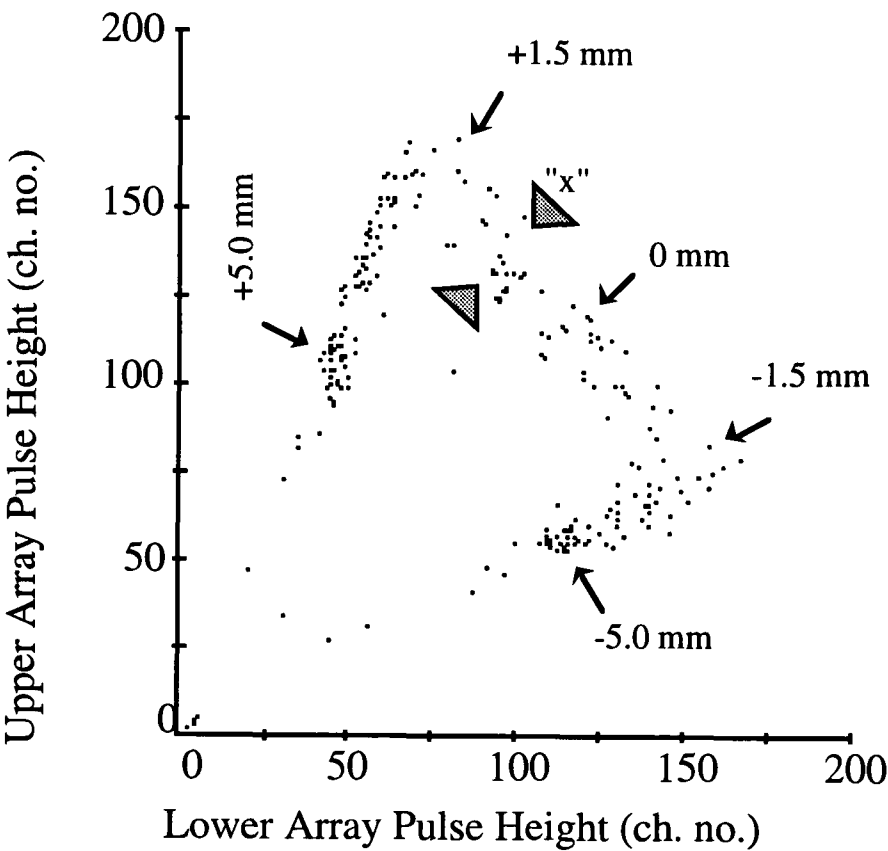


Figure 7.3.1 Peak voltage signals from the upper versus lower SASTJ on the face opposite to incident 5.5 MeV α -particles (See inset of Fig. 7.3.2).

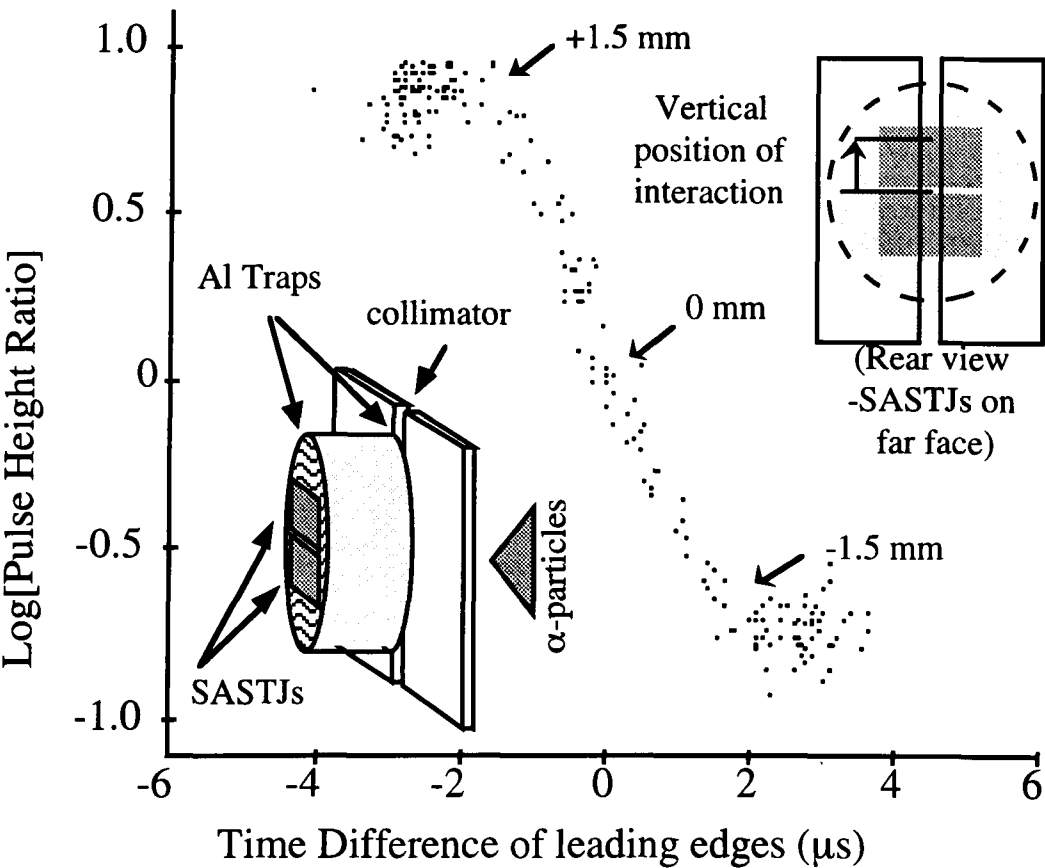


Figure 7.3.2 A plot of the time difference of the leading edges of the signals from the two arrays versus the log of the ratio of the pulse heights. The Nb crystal was 3 mm length x 10 mm diameter and the arrays cover an area of 2.9 x 6 mm each with a centre separation of 3 mm.

The width of the scatter distribution (marked “x” in Fig. 7.3.1) is greater than that which would be observed in experiments on crystals where the α -particles are incident on a bare surface of the Nb. The presence of an Al trap on the Nb crystal surface in the immediate vicinity of the α -particle interaction region appears to affect the initial evolution of the non-thermal signal.

A plot of the time difference of the leading edges from the upper and lower arrays is shown in Fig. 7.3.2. The distribution implies a propagation speed of 1.0 km s^{-1} compared to the slow transverse phonon velocity of $\sim 2 \text{ km s}^{-1}$. Once the interaction takes place outside the central region, the timing difference becomes weakly position dependent as would be expected. The position resolution for the interactions on the far face is $\sim 200 \text{ }\mu\text{m}$, as ascertained from the signal timing differences in the two arrays.

Knowledge of the position dependence of the α -particle signals for this device aids the interpretation of the signals arising in the SASTJs for γ -ray interactions where the position cannot be determined a priori. The device was used to detect 662 keV and 356 keV γ -rays from sources outside the cryostat. Figure 7.3.3 shows a plot of the pulse heights from the upper and lower SASTJs (connected in series as indicated in the inset of Fig. 7.3.4) for 662 keV γ -rays from ^{137}Cs source. The reduced position dependence shown in the scatter plot of Fig. 7.3.3 compared to Fig. 7.3.1 indicates a difference between the shape of the signals arising from α and γ -ray interactions ¹⁶⁶. (We would expect the altered array connections to accentuate the position dependence.) A different behaviour is also observed in the range of the timing differences between the leading edges in Fig. 7.3.2. More detailed modelling of the signal shapes and position dependencies is currently being carried out.

¹⁶⁶At present this is only verified statistically by the scatter of the points.

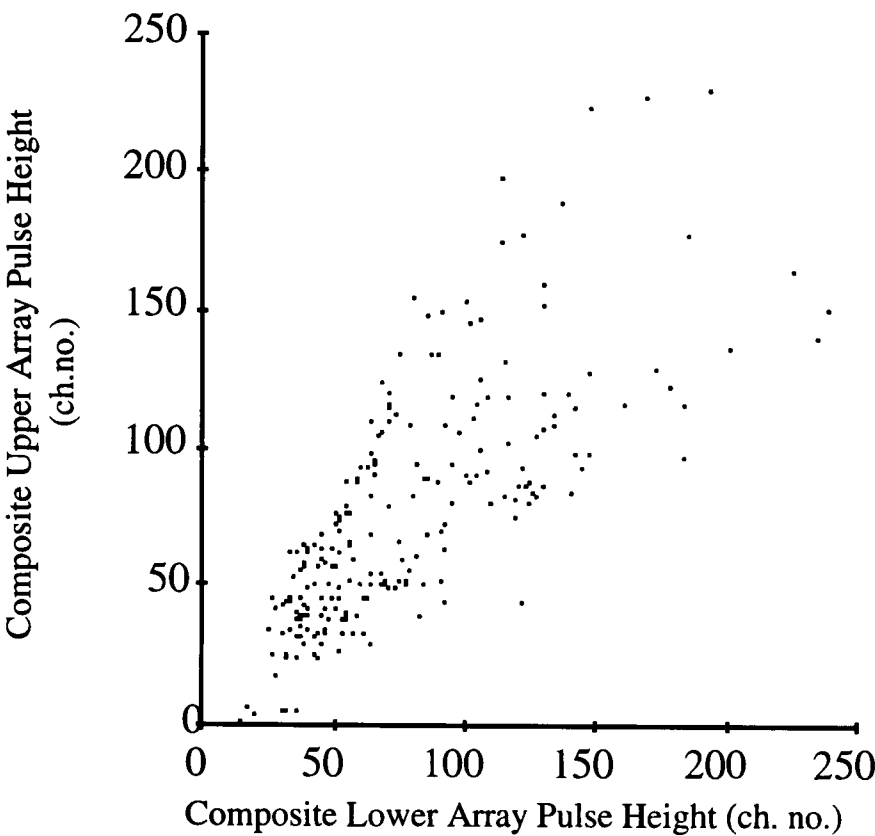


Figure 7.3.3 Voltage pulse heights in upper and lower composite arrays. On the same device as shown in Fig. 7.3.2, the arrays were paired so that those opposite one another were connected in series (as shown in inset of 7.3.3). The device was irradiated with 662 keV γ -rays from outside the cryostat.

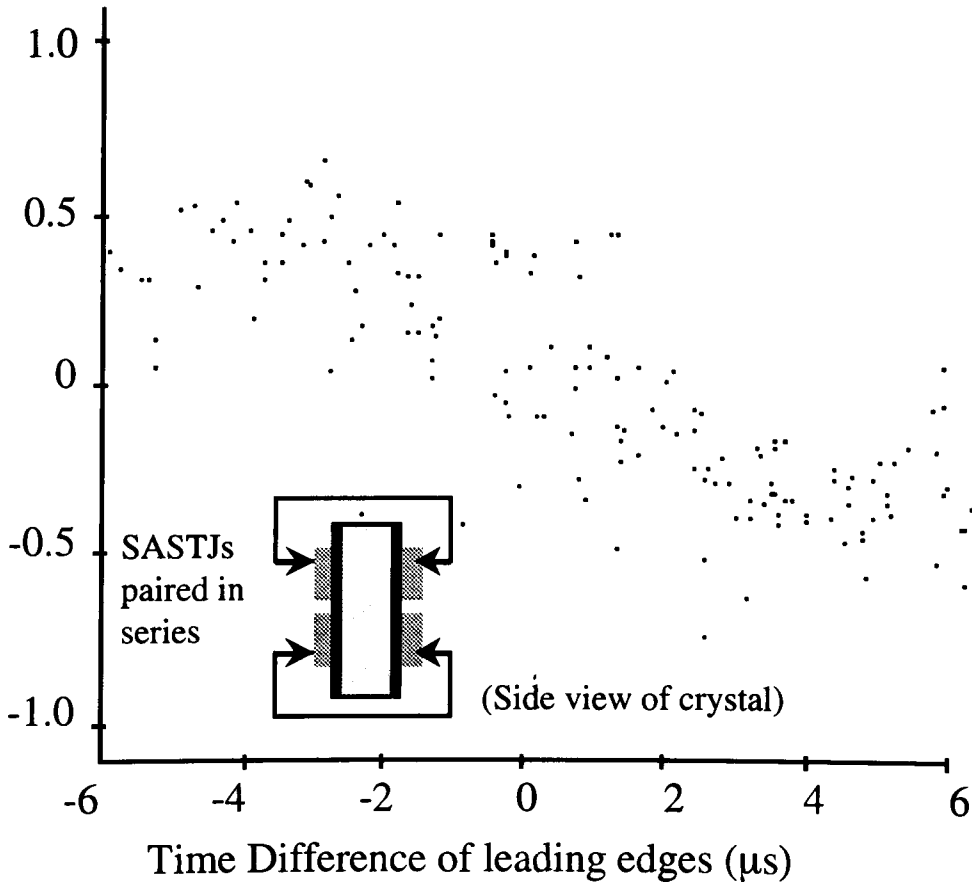


Figure 7.3.4 The timing difference of the rising edges of the signals in the upper and lower composite arrays (connected as shown in the inset) for irradiation by 662 keV γ -rays from outside the cryostat.

We would expect that, at a time long compared with (a), the initial propagation time of the signal in the Nb crystal and (b), the integration time of the Al SASTJs determined

by the quasiparticle recombination time in the Al thin films, the signals will become less position dependent. Comparison of the signal magnitudes at $> 100 \mu\text{s}$ in two separate SASTJs on the 2 g crystal confirms this. Figure 7.3.5 shows a pulse area spectrum obtained by integrating the signal in the window $100\text{--}160 \mu\text{s}$ for $356 \text{ keV } ^{133}\text{Ba}$ γ -rays from a source external to the cryostat. Though with limited statistics, the spectrum is consistent with the observation of a 356 keV photoelectric peak.

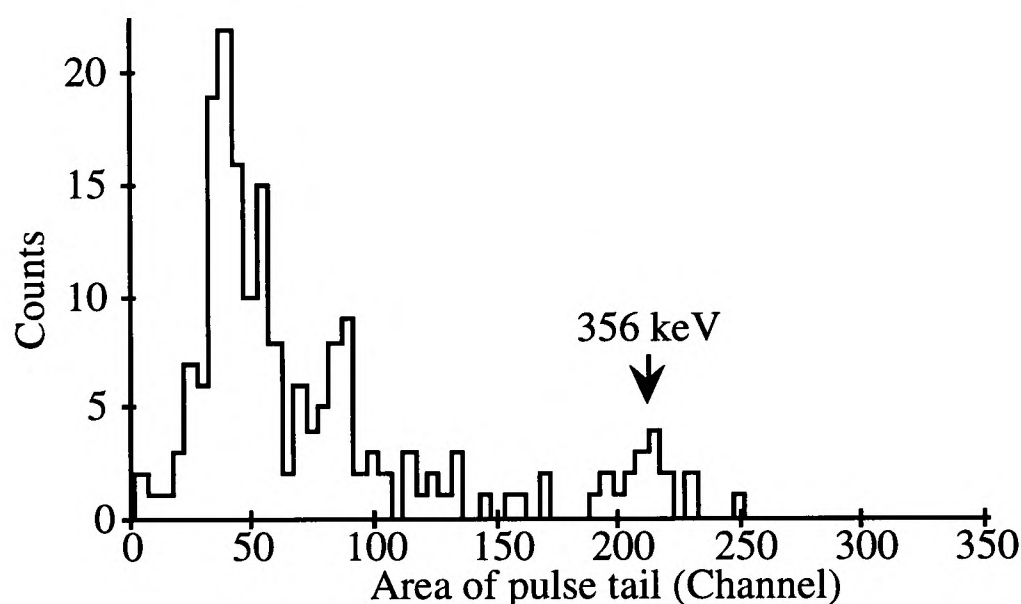


Figure 7.3.5. A spectrum of the pulse tail area for a signal in an SASTJ on the device shown in Fig. 7.3.4 irradiated with ^{133}Ba γ -rays from a source external to the cryostat.

7.4. PARTICLE DETECTION EXPERIMENT IN A DILUTION REFRIGERATOR

An Oxford Instruments dilution refrigerator has recently been commissioned at the Oxford Cryodetector group. A preliminary investigation was made of α -particle detection at a temperature of 70 mK using the device discussed in §8.2. The results were surprising.

The 12 g Nb crystal was irradiated with α -particles using an identical collimation to that shown in Fig. 7.2.1. The voltage pulses from the left and right arrays at a temperature of 70 mK for an interaction in the far left hand hole are shown in Fig. 7.4.1 (compare with Fig. 7.2.2). The voltage signal decay times of the pulses have become comparable with our estimate of the recombination time in the Al thin films ¹⁶⁷. This implies that the above-gap phonon flux entering the SASTJs must have very short lifetimes. The ratio of

¹⁶⁷We were unable to check the recombination time of the quasiparticles in the Al films directly since no laser fibres were installed in the dilution refrigerator.

the heights of the voltage signals in the left and right arrays has also become more exaggerated. The signal heights now appear to scale approximately with the square of the propagation distance. This is the behaviour we would expect if we were only detecting the ballistic component of the phonon signal.

While this result is only preliminary, if correct it implies the decay of phonons at the surface of the Nb takes place more rapidly than we had imagined. The results obtained using an Al–Ox–Al STJ, at 360 mK, reported in §5.5 indicated a decay time of 145 μs for the phonon signal in the range 50–150 μs . This would have seemed to be close to a limiting value since at this temperature the thermal population of phonons above 50% of Δ_{Al} is already becoming small (see Fig. 5.5.13). Phonons in this energy region would be expected to make a significant contribution to the 3–phonon processes that up–convert non–equilibrium phonons that have decayed and so prolong their apparent lifetime. We intend to study this effect in more detail.¹⁶⁸

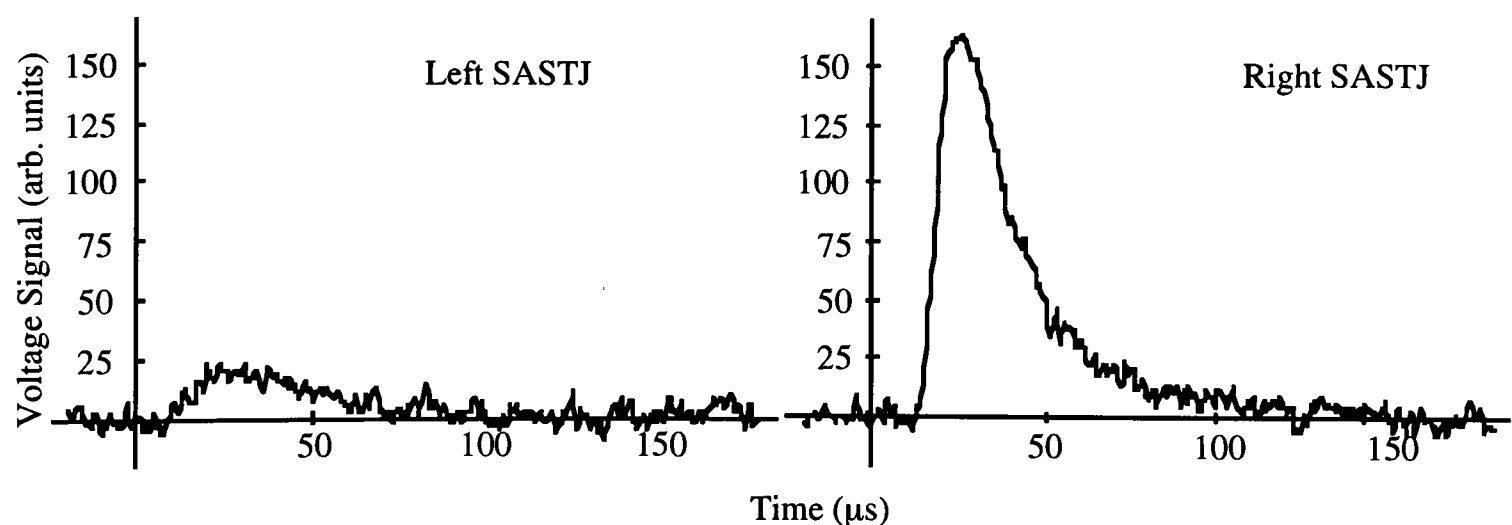


Figure 7.4.1 Voltage signals from constant current biased SASTJs at opposite ends of the crystal following an α –particle interaction in the hole nearest the right hand end. The pulses were recorded in a similar experimental arrangement to used to record those shown in Fig. 7.2.2 except that the temperature is now 70 mK.

¹⁶⁸Investigations subsequent to the completion of this work indicate that the pulse shapes of the signals shown in Fig. 7.4.1 recorded from Nb crystals in the dilution refrigerator are anomalous. In subsequent experiments the pulse shapes have shown decay lifetimes in excess of those illustrated in Fig. 7.4.1 by a factor of 2–5. We believe that this effect could be caused by the inclusion of magnetic flux in the Nb crystal and arrays while cooling the samples. We are currently investigating this and related phenomena.

7.5. CONCLUSION

We have demonstrated the energy and position resolution of the 12 g Nb detector for 5.5 MeV α -particles to be 170 keV and ~ 500 μm FWHM respectively. The threshold of the detector at 360 mK is limited by the electronic noise. A significant improvement in the sensitivity of the SASTJs is expected at lower temperatures.

Initial work with 2 g Nb crystals has shown a difference in the propagation characteristics of signals arising from α -particle and γ -ray interactions. We will investigate this phenomenon further and also look for differences in signals arising from nuclear recoil events.

The initial results from the dilution refrigerator experiment were surprising. It is clear that further investigations of the behaviour of non-equilibrium phonons in Nb will have to be conducted if we are going to be able to realise a large mass Nb particle detector.

8. CONCLUSIONS

*Do not all charms fly
At the mere touch of cold philosophy ?
There was an awful rainbow once in heaven:
We know her woof, her texture; she is given
In the dull catalogue of common things.
Philosophy will clip an angel's wings,
Conquer all mysteries by rule and line,
Empty the haunted air, and gnomed mine
Unweave a rainbow.*

John Keats

We have observed the ballistic propagation of phonons in high purity Nb at temperatures below 1 K. The propagation velocities agree with calculations based on the tabulated second-order elastic constants. Some evidence for the predicted anisotropy of the phonon propagation times was observed. At higher temperature ($T \geq 2$ K) it was shown that the phonon m.f.p. becomes limited by scattering from the thermally generated quasiparticles. The characteristic scattering times were found to agree with theory.

The behaviour of the reverberation of non-equilibrium phonons was unexpected. The lifetimes of non-thermal phonons were very much shorter than estimates based on the bulk anharmonic decay rates. We believe significant down conversion was taking place at the surface of the crystal. The down conversion rates were strongly temperature dependent. The size of the signal at long time scales was found to be close to that

predicted for a fully thermal phonon distribution, assuming the heat capacity was given by the Debye model. The signal decay time was found to correspond to the thermal time constant of the crystal.

Quasiparticle propagation was observed in junctions fabricated directly on the Nb crystal surface. We were unable to unambiguously identify the quasiparticle signal in junctions separated from the crystal by an insulator.

Initial work to investigate quasiparticle trapping into thin films was frustrated by poor interfaces between the Nb crystals and the Al trapping films. Better conditions have now been identified for trap fabrication. We will investigate trapping again shortly.

α -particle and γ -radiation interactions were observed in bulk Nb crystals using Al SASTJs. We demonstrated methods for extracting the position and energy of the interaction. Differences were found in the behaviour of the non-thermal signals arising from α -particle and γ -ray interactions. In a 12 g Nb crystal at 360 mK the detection threshold was ~ 200 keV. We plan to investigate the improvement in threshold (due to enhanced SASTJ sensitivity) when the device is cooled to lower temperatures.

9. REFERENCES

It may well wait a century for a reader, as God has waited six thousand years for an observer.

Johannes Kepler

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10. APPENDICES

A. MEASUREMENT OF QUASIPARTICLE RECOMBINATION TIME IN SUPERCONDUCTING THIN FILMS

In this appendix we show the results of the direct measurement of the recombination times in the STJ and SASTJ used in §5.5. The times were measured from the shape of the voltage pulses in the devices following illumination with direct laser light. For the low temperature points we fitted the pulses using the Eq. 3.4.9 using a value for τ_{RC} determined from the dynamic resistance of the device and an estimate of the total capacitance of the device plus electronics and wiring. The high temperature points for the Sn device were obtained by a measurement of the voltage pulse area ($\sim\tau_r$).

The apparent enhancement of the quasiparticle lifetimes over those calculated using the value for τ_0^{qp} in Table 3.3.1 and Eq. 3.3.2 is due the recombination phonons breaking Cooper pairs before they are able to leave the film (see §3.3.3). The phonon trapping factor F , which is the ratio of the observed recombination time and the calculated value (divided by 2) can be related to the phonon loss time τ_γ from the films by

$$F = 1 + \frac{\tau_\gamma}{\tau_{pb}}, \quad [A.1]$$

where τ_{pb} is the calculated pair breaking time in the films according to Eq. 3.3.22. The calculated phonon loss times for the Al SASTJ and Sn STJ are 400 ps and 1 ns. This consistent with those values quoted in Eisenmenger. [60, 143].

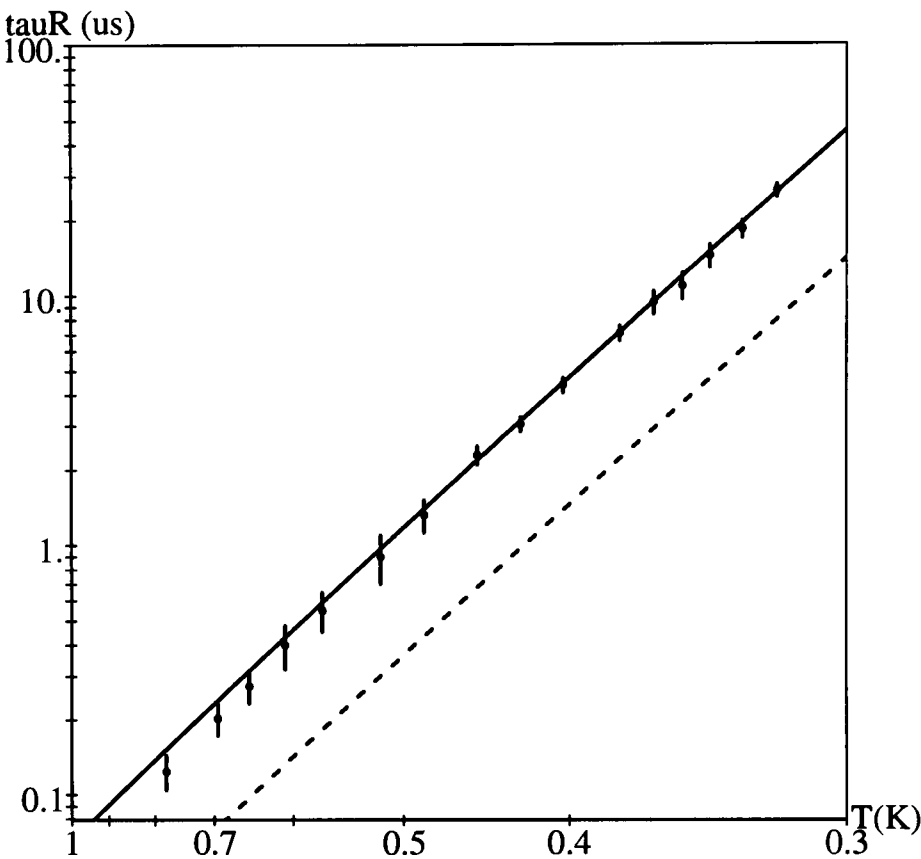


Figure A.1 Recombination time of quasiparticles in Al film of Al SASTJ (§5.5.7). Dotted line is (half) calculated recombination time using $\tau_0^{qp} = 110$ ns. Experimental recombination times were extracted from direct illumination pulses and fit using a convolution algorithm to take account of electronic RC effects. The superconducting energy gap used in the fit was $\Delta = 220 \mu eV$.

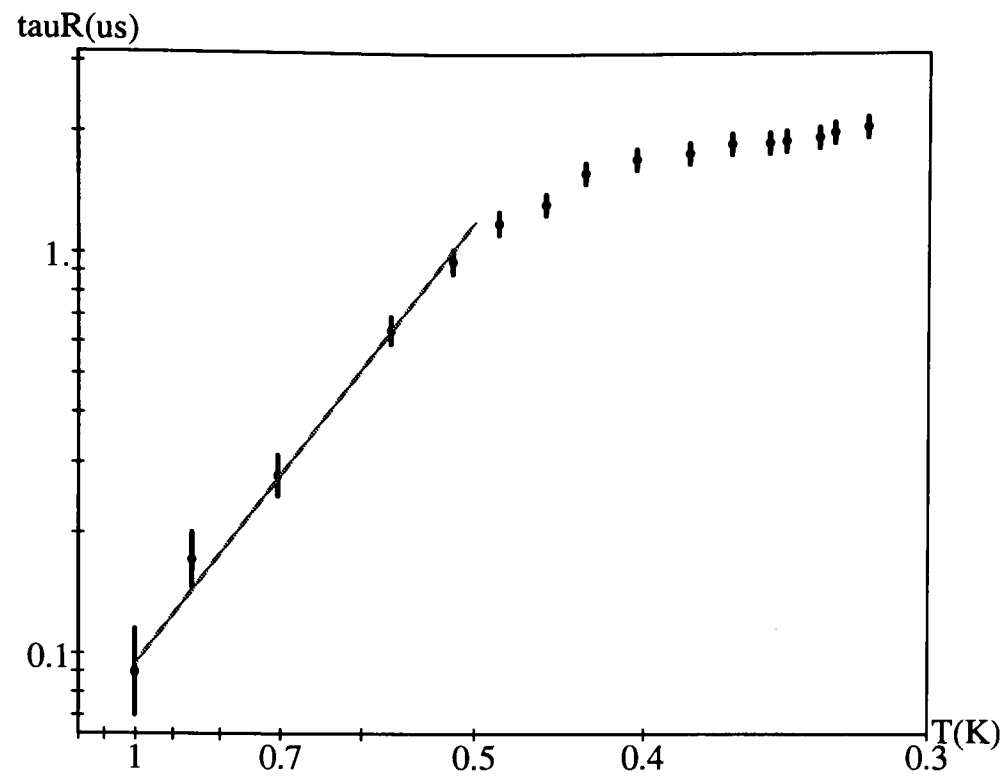


Figure A.2 Al recombination time in STJ (§5.5.2). The saturation at low temperatures was caused by self-heating. The line gives a slope of $220 \mu\text{eV}$.¹⁶⁹

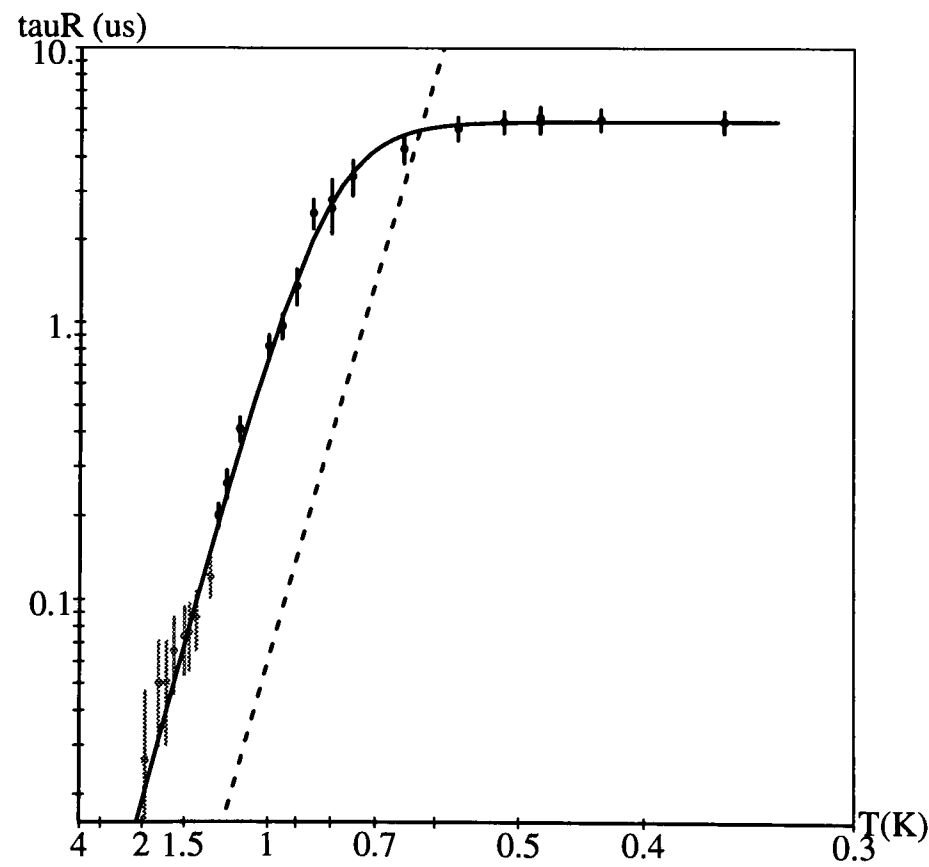


Figure A.3 Sn film recombination time as a function of temperature (§5.5.3). High temperature data in grey was obtained by measurement of the area of the direct signal in order to avoid RC effects. The fit uses a slope of $600 \mu\text{eV}$ saturating at $5.4 \mu\text{s}$.[†]

¹⁶⁹File: "nb140793 VIIIp71 Pulse Series"
[†] File: "TauR Final Ver 4.11.92 VIIIp104"

B. EARLY MODEL EXPLAINING TEMPERATURE DEPENDENCE OF SIGNAL ARISING FROM QUASIPARTICLES IN BULK NIOBIUM

We discuss briefly an alternative model for the tail signal discussed in terms of the thermalisation of phonons in §5.5. In this case the signal arises from the anomalous recombination of quasiparticles. The fit to the data is almost as good as that from the phonon model, however, the arbitrary parameters included in this second model do little to recommend it.

If the tail signal (at long times) discussed in §5.5 had arisen from quasiparticle recombination in the bulk of the crystal we would expect the phonon flux to vary as $e^{-\Delta_{Nb}/k_B T}$. In the cases of the Sn STJ and Al SASTJ were observed the tail flux to show an approximate temperature dependence $e^{-\Delta_{film}/k_B T}$ where Δ_{film} is 600 and 220 μeV respectively. The precise mechanism of recombination would have to be tailored to explain the observed temperature dependence of the signal flux. We compare the predictions of the two models in Fig. B.1.

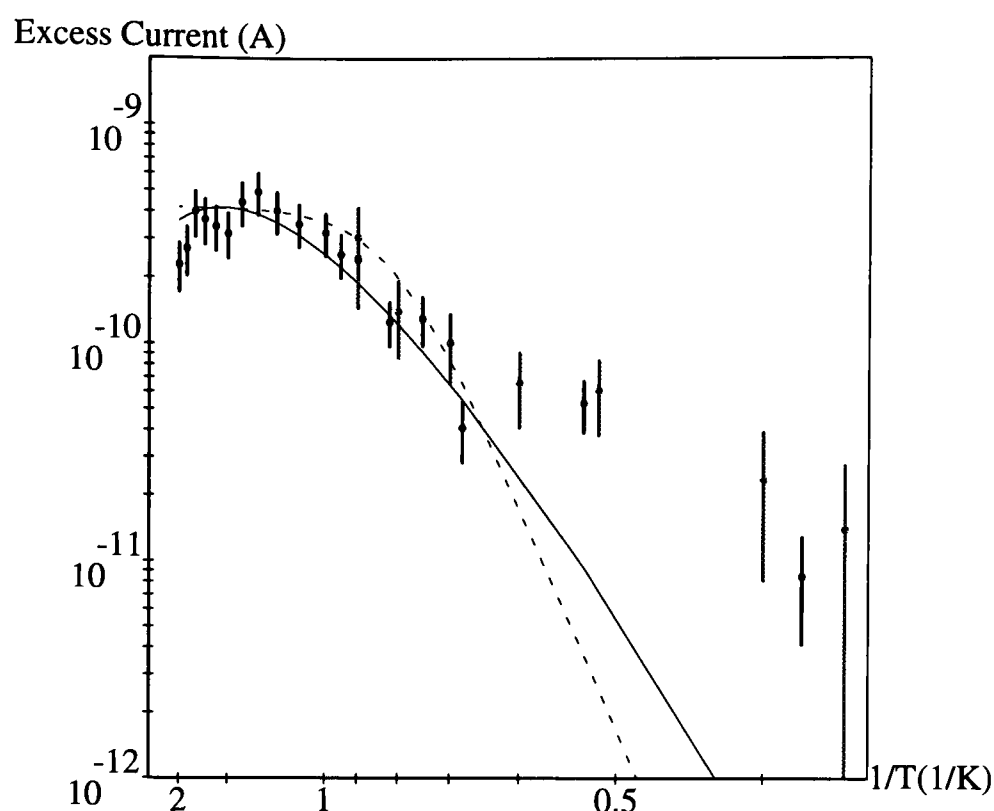


Figure B.1 Comparison of excess signal current in Sn-O_x-Sn STJ with temperature against predictions from phonon (solid line) and quasiparticle (dashed line) models. The absence of free parameters in the phonon model favours this interpretation.^{†1}

If the quasiparticles were predominantly recombining, not in the bulk of the crystal, but at the surface of the crystal in a region where the superconducting energy gap was reduced then it is possible to suggest a reason for the observed temperature dependence. The regions where the gap is reduced would have thermally generated quasiparticle populations dependent of the local value of the gap. Excess quasiparticles from the bulk of the crystal would be able to recombine in these regions emitting phonons. The number of phonons emitted ($\Omega = 2\Delta_{\text{Reduced}}$) would be proportional the thermal populations at a given reduced gap Δ_{Reduced} . We would assume that the dominant phonon component at a given threshold came from recombination of quasiparticles at that threshold. The relaxation phonons emitted by the quasiparticles as they moved into this region would have to be very low energy (so as not be detected by STJ). This could arise if the depth of such a damage region was large compared to the superconducting coherence length.

^{†1} File: "Phonon T3 vs QP Recomb Sig 12.1"

C. STJ DETECTOR RESPONSE FUNCTION

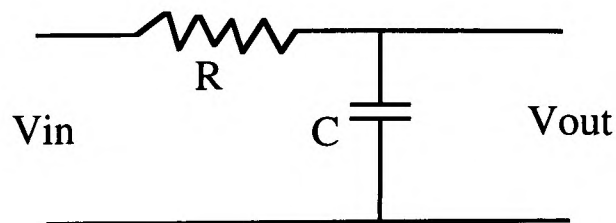


Figure C1 RC filter

The basis of Eq. 3.4.9 is the convolution of the effect of an RC filter which is described by the following equation:

$$v_{out}(t) = \frac{1}{RC} \int_{-\infty}^t v_{in}(t') e^{\frac{-(t-t')}{RC}} dt'$$

with an input signal (which corresponds to the injection of quasiparticles at $t=0$ and their subsequent decay due to recombination),

$$v_{in}(t) = e^{\frac{-t}{\tau_{Decay}}}$$

The solution has the form

$$v_{out}(t) = \frac{\tau_{Decay}}{RC - \tau_{Decay}} \left(e^{\frac{-t}{RC}} - e^{\frac{-t}{\tau_{Decay}}} \right).$$

In the three cases of the relative sizes of the time constants this is

$$\tau_{Decay} = RC, \quad v_{out}(t) = \frac{t}{RC} e^{\frac{-t}{RC}}$$

$$\tau_{Decay} \gg RC, \quad v_{out}(t) = e^{\frac{-t}{\tau_{Decay}}} \text{ (original signal)}$$

$$\tau_{Decay} \ll RC, \quad v_{out}(t) = \frac{\tau_{Decay}}{RC} e^{\frac{-t}{RC}}$$

