# Transition-Edge Sensors for X-ray Astronomy

Thesis Submitted for the degree of Doctor of Philosophy at the University of Leicester

by

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May 16, 2001

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# **Transition-Edge Sensors for X-ray Astronomy**

### **Stephen P. Trowell**

## Abstract

This thesis considers the development of a cryogenic Transition Edge Sensor (TES) microcalorimeter for X-ray astronomy. An optimum configuration for this application has yet to be established and some of the design criteria and fabrication methods are discussed in Chapter 5. Results from investigations into different TES material combinations are presented in Chapter 6. Degradation due to interdiffusion between materials has an impact on the reliability of TES detectors and analysis of Al/Ag, Ti/Au, Cu/Nb and Ir/Nb systems has formed a large part of our recent work. Such comparisons show the relative stability of iridium-based detectors. The superconducting properties of thin film iridium samples were investigated in the context of detector development. The measured superconducting-to-normal transitions are presented in Chapter 7. Changes in transition temperature from 157 mK to 52 mK were observed. A model based on the formation of thin iridium silicide layers during film deposition is proposed to describe this effect. Chapter 8 contains results from testing of a Ti/Au single pixel TES device. The capability of our digital signal processing system was demonstrated with the acquisition of X-ray spectra with photopeak energy resolutions of 25 eV FWHM at 6 keV, close to the detector limit. Imaging spectrometers are required for astronomy applications and our extended bismuth absorber with distributed read-out concept is described in Chapter 9. Phonon propagation through bismuth at low temperatures (~100 mK) has been modelled using a Monte Carlo approach, and the results of the simulations indicate that large square bismuth absorbers (of side length ~5 mm) with corner TES read-out can achieve spatial resolutions better than 250 µm. This is adequate to satisfy the current specification for the narrow-field imaging spectrometer instrument of ESA's proposed XEUS mission.

# Declaration

I hereby declare that no part of this thesis has been previously submitted to this or any other university as part of the requirement for a higher degree. The work described herein was conducted solely by the undersigned except for those colleagues and other workers acknowledged in the text.

Syn Tra

Stephen P. Trowell May 16, 2001

# **Publications**

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# **Dedication**

This thesis is dedicated to my Mum, Dad and Nic, in thanks for their endless love and support.

# Acknowledgements

This thesis would not have been written if it were not for the support of many individuals, a list too numerous for me to thank them all by name here. Hopefully, those not explicitly mentioned below know how grateful I am for their input.

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# **Chapter 1**

# Introduction

### 1.1 Cryogenic Detectors for X-ray Astronomy

Since the discovery of the first cosmic X-ray source in June 1962 [Giacconi, R., 1962], X-ray astronomy has developed into one of the major fields of astrophysics. Observations in the X-ray band provide the only means to directly observe hot plasmas and matter under extreme temperature, pressure, density and intense gravity conditions. Hence, X-ray astronomy has a unique potential for understanding the distant universe and has profound implications for cosmology and the life cycle of matter.

The Earth's atmosphere is an effective absorber at X-ray wavelengths and so astrophysical observations are restricted to satellite based platforms (or high altitude balloons for E > 30 keV). The current generation of space-borne X-ray telescopes include Chandra (NASA) [Weisskopf. M., 1987], XMM-Newton (ESA) [Lumb, D., 1996] and the Astro-series (Japan) [Ogawara, Y., 1998 and Inoue, H., 1995]. Each of these offers significant advances in collecting power and angular and spectral resolution over previous missions such as ROSAT and support a large international observer community. The present 'state of the art' in high resolution X-ray spectroscopy is defined by instruments such as the Low Energy Transmission Grating Spectrometer (LETGS) on Chandra [Zombeck, M., 1995]. This is capable of a resolving power,  $E/\Delta E > 1000$  when used in conjunction with the High Resolution Camera Spectroscopic array (HRC-S) in the energy range 0.07 – 0.15 keV, but suffers from the low throughput of any transmission grating (~10%). Other examples of current technology include Charge Coupled Devices (CCDs) such as the European Photon Imaging Camera (EPIC) on XMM [Holland, A., 1996] with  $E/\Delta E \sim 10$  at 1 KeV and the Reflection Grating Spectrometer (RGS), also on XMM [Brinkman, A., 1996].

In order to continue the expansion in observational capabilities, there is widespread agreement within the X-ray astronomy community that the next generation of X-ray observatories should concentrate on high resolution spectroscopy [XEUS Science Case, 2000]. There are strong scientific drivers for high throughput photon-counting (high quantum efficiency) instrumentation with resolving powers,  $E/\Delta E >$ 1000 (see section 1.2.1) and several groups around the world are investigating the new detector technologies required to meet these demands. At present, two main cryogenic detector technologies exist with the potential performance for future missions. At Leicester University, we have concentrated our efforts on Transition Edge Sensor (TES) microcalorimeters, which we believe show greatest promise for the future. In a microcalorimeter, the thermal energy from an absorbed X-ray photon is measured and used to deduce the energy of the incident photon. In a TES, the thermometer is the changing resistance of a superconducting layer biassed on its superconducting-to-normal transition. A group at the National Institute of Standards and Technology (NIST), Colorado has achieved the best performance to date for a TES: 2.0 eV FWHM at 1.5 keV [Wollman, D., 2000]. This thesis is concerned with the development of a TES to meet the performance requirements of a narrow field instrument for the proposed XEUS mission [Bavdaz, M., 1998]. Such detector technology also has applications in ground-based spectroscopy for materials analysis (section 1.3).

The main alternative cryogenic detector technology under consideration is the Superconducting Tunnel Junction (STJ). In these devices, a photon is absorbed in a superconducting layer, typically niobium, tantalum or hafnium, generating hot electrons [Kraft, S., 1998a]. These thermalise by excitation of phonons which can break the Cooper pairs and create a large number of charge carriers (quasiparticles). The quasiparticles are detected as a current pulse arising from tunnelling across an insulating layer, often aluminium oxide, in the STJ. The initial number of quasiparticles created is proportional to the energy of the absorbed photon and, theoretically, the energy resolution is determined by statistical variations in the number of quasiparticles created (2.4 x  $10^6$  for a 6 keV X-ray absorbed in niobium) and in the back-tunnelling process, or Gray effect [Gray, K., 1978]. On average, a single quasiparticle in a tantalum device tunnels 10 times across the thin oxide barrier [Kraft, S., 1998b]. For a symmetrical junction, the intrinsic energy resolution,  $\Delta E$  scales as the square root of energy [Rando, N., 1998a]:

$$\Delta E(eV) = 2.35 \sqrt{\varepsilon E \left(1 + F + \frac{1}{\langle n \rangle}\right)}$$
(1-1)

Where  $\varepsilon$  is the mean Cooper pair binding energy (~1.2 meV for Ta), *F* is the Fano factor (~0.2 for elemental superconductors), and  $\langle n \rangle$  is the average number of times each quasiparticle tunnels before recombining. From equation (1-1), as  $\langle n \rangle \rightarrow \infty$ ,  $\Delta E \sim 7$  eV at 6 keV. In practice, at X-ray energies, energy resolution is limited by spatial non-uniformities in responsivity, which is not the case at lower energies [Poelaert, A., 1998]. At optical wavelengths, the signal is small and energy resolution is limited by the signal to noise ratio [Rando, N., 1998b]. In optical high-resolution spectroscopy, a resolving power,  $\lambda/d\lambda > 10^4$  is typically required. This implies a transition temperature of  $\langle < 100 \ \mu K$  [Peacock, A., 1998]. Therefore, applications of tantalum-based STJs in optical/UV astronomy are restricted to low energy resolution observations that require accurate timing information ( $\langle 10 \ \mu s$ ) or high quantum efficiency.

Tantalum based STJs have been developed which reach their theoretically predicted resolving power in the optical band [Verhoeve, P., 1997], but in the X-ray band (E > 1 keV) additional charge losses degrade the detector response. There are also constraints on the device thickness set by fabrication (<500 nm for tantalum) which limits detection quantum efficiency to < 20 % at 6 keV [Kraft, S., 1998b]. Despite these limitations, STJ development has progressed further than that of TESs and a tantalum-aluminium based device has been fabricated as a 6 x 6 imaging array [Rando, N., 1998a]. Pixels in this array have measured energy resolutions of 6.2 eV – 7.9 eV at 1 keV, with a quantum efficiency of 30 % and an operating temperature of 400 mK [Verhoeve, P., 1999].

STJs will undoubtedly provide a significant additional tool for astronomy in the next century and one is currently included in the baseline design for XEUS (section 1.2). However, in contrast to STJs, TESs have lower theoretical limits on energy resolution for X-rays above 1 keV (where STJ quantum efficiency is poor) and, in the case of a normal metal absorber, simpler relaxation processes after photon absorption. Quasiparticle loss and trapping mechanisms within superconducting absorbers are complex and are currently limiting the energy resolution achievable with an STJ. Therefore, it is our belief that TESs currently offer the greatest potential for high resolution X-ray spectroscopy.

### **1.2 XEUS**

The X-ray Evolving Universe Spectroscopy mission (XEUS) is a European concept to succeed the XMM-Newton cornerstone mission that was launched in December 1999. The XEUS mission concept was initially conceived in July 1996 during discussions of the European, US and Japanese Scientific Communities at an international workshop on the next generation of X-ray observatories, held at Leicester University [Turner, M., 1997]. The XEUS mission aims to elevate X-ray astronomy to the level of other wavelength disciplines by placing a telescope aperture in space equivalent to the largest ground-based optical telescope built to date. In the final configuration, a XEUS mirror with a diameter of 10 m is planned [Willingale, R., 1997]; necessary to study the distant, early universe. This compares to the XMM mirror module diameter of 0.7 m and the 1.3 m diameter mirror assemblies of the proposed US mission, Constellation-X [Weaver, K., 1999]. Extensive use will be made of the facilities of the International Space Station (ISS) to enable significant growth and evolution of the XEUS observatory, resulting in the very large optic and a possible operational lifetime of over 25 years. Such a large aperture yields high photon throughput: approximately 25 times that of XMM. This places stringent requirements on the speed of operation for the detectors. Such a high flux is necessary to fill the spectral bins of high energy resolution instrumentation and for the first time will allow the full exploitation of cryogenic detector technology.

### **1.2.1 XEUS Science Goals**

Four key scientific drivers can be identified for XEUS [XEUS Science Case, 2000]. Firstly, there is the exploration of the distant and evolving high energy universe. This includes the study of the formation and evolution of galaxies and clusters and the evolution of massive black holes in Active Galactic Nuclei (AGN). Such studies of the early universe will complement the lower energy waveband investigations proposed for the Next Generation Space Telescope (NGST). Another driver is the study of the behaviour of matter under strong gravity i.e. near the event horizon of black holes. This can provide detailed characterisation of black holes as well as direct tests of general relativity in the strong gravity limit. Thirdly, the creation, redistribution and life cycles of the chemical elements is to be investigated. Finally, the open question of the distribution of dark matter can be addressed by measurement of the temperature and density profiles of hot gas out to large radii from clusters of galaxies where it is believed most of the dark matter resides.

### **1.2.1.1 High Resolution Spectroscopy**

Much of the expected science yield from XEUS will come from the narrow field instruments, for which a TES is a possible candidate. With a TES, the physical parameters of the emitting regions of all classes of X-ray emitters may be probed; including stars, supernovae remnants, X-ray binaries, galaxies and clusters of galaxies, quasars and interstellar and intergalactic material. Many of these sources are at high redshift, therefore the lines and edges move to lower energies. For z = 4, energies of 0.5 keV – 2 keV in the rest frame are transformed into observations between 100 eV – 500 eV. The desire to resolve such lines is a key science driver for high spectral resolution instrumentation on XEUS.

### **1.2.1.2 Plasma Diagnostics**

The TES energy range (0.1 - 10 keV) is rich in emission line plasma diagnostics and this is a likely source of new astrophysics. Through study of the emission lines, absorption lines and absorption edges it is possible to determine source properties such as temperatures, densities and relative abundances, velocities and ionisation states [Paerels, F., 1997]. For example, the temperature and density of a plasma can be diagnosed from the line ratios of forbidden, intercombination and resonance lines of the He-like ions [Mewe, R., 1997]. Electron temperature may be determined using satellite line spectroscopy. Satellite lines of an ion arise due to the shielding effect of a 'spectator' electron in a high Rydberg level. The ratio of the intensities in the 'parent' and satellite lines depends on the collisional excitation rate and dielectronic recombination rate, both of which vary with electron temperature [Liedahl, D, 1997]. Beiersdorfer et al. [1992] conclude that a resolving power > 1000 is required to resolve the n=3 satellite line of the Fe resonance line at 6.701 keV.

The majority of the diagnostics lie below 2 keV where line blending is pervasive: in solar spectra taken with a resolution of 400, spectral bins contain contributions from up to 6 lines [McKenzie, D., 1980]. Unambiguous decomposition again demands a minimum resolving power of 1000. Both STJs and TESs are theoretically capable of this performance but currently neither have demonstrated such energy resolution, and it is unclear which technology may prevail for this application.

### **1.2.1.3 Doppler Motion**

Spectroscopy also yields Doppler velocities. For kT = 1 keV, a medium mass ion has a thermal velocity of ~ 100 kms<sup>-1</sup>. This is a typical value for Doppler motion in binary systems, stellar winds, spiral galaxies, and mergers of clusters of galaxies [Elvis, M., 1997]. Doppler variations of this order in the (6.40 keV) Fe line in X-ray binaries can allow the masses to be determined directly. However, in practice resolutions of better than 1000 are required, particularly if two velocity components are blended which is likely to be the case in many systems.

### **1.2.1.4 Protogalaxies**

Theory predicts that at high z there should be many protogalaxies which become less common toward the present, forming the galaxies we see around us [Wolfe, A., 1995]. Likely candidates for these protogalaxies are the damped Lyman- $\alpha$  systems that have been observed in optical/UV measurements of non-luminous material toward quasars. X-ray K-edges give more accurate estimates of abundances and ionisation states than saturated optical/UV lines of individual ions [Madejski, G., 1995]. The hot Interstellar Medium (ISM) can be effectively probed using X-ray absorption lines which can be studied at 1000 times smaller column density than absorption edges. This allows the study of the 'metal line systems' that lie around developed galaxies [Steidel, C., 1995].

### 1.2.1.5 Supermassive Black Holes in Active Galactic Nuclei

Fe-K lines can be used to probe the unresolved, compact central engine of Active Galaxies. ASCA observations [Tanaka, Y., 1995] show broad Fe-K lines originating from close to the black hole event horizon, probably from the inner edge of the accretion disk. This broadening can be understood as a combination of Doppler shift and gravitational redshift and provides the strongest evidence to date for the existence of supermassive black holes in the centres of Active Galaxies. Missions such as XMM facilitate measurement of the detailed profile of the iron line, which can be used to measure properties of the black hole such as its spin-rate.

High throughput spectroscopy missions such as Constellation-X or XEUS are required to make further progress via the search for iron line reverberation (correlated variability between the line and the X-ray flares which drive it) [Reynolds, C., 1999]. If detected, iron line reverberation should allow us to map out the structure of material as it falls into the central black hole. These measurements will provide the best probe of gravity in the strong-field regime and allow new tests of General Relativity.

### 1.2.1.6 Stellar Winds

Stellar mass loss contributes to the chemical evolution of the Galaxy by enrichment of the ISM. OB stars have mass flows of around 1000 kms<sup>-1</sup> and so with a spectral resolving power of 1000 it is possible to measure Doppler broadening and line profiles of X-ray lines emitted in the wind. The results can be used to test models in which the X-ray emission is thought to arise from shocked wind material.

### **1.2.2 XEUS Design Features**

Designs are at an early stage of development but the current concept is to fly two spacecraft – a mirror module containing the optic and a second spacecraft containing the focal plane instrumentation. This unique configuration will establish the required 50 m focal length without the need for a long stabilising tube which would be difficult to launch and deploy. Flying the two spacecraft to a relative accuracy of 1 mm<sup>3</sup> is within modern attitude control capabilities. Another key feature of XEUS is the large aperture X-ray mirror. This will derive from the mirror technology developed in association with industry for the XMM programme. The XMM design used heavily nested mirrors fabricated from closed shells. In contrast, the XEUS aperture is divided into annuli with each annulus sub-divided into sectors. Each free–standing sector has a target angular resolution of ~ 2 arcsecond and an energy range of 50 eV to 30 keV. At launch (around 2015) the optic will consist of only the two inner annuli giving an effective area of 5 m<sup>2</sup> with a diameter of 4.5 m. The outer annuli will be added in orbit using the ISS facilities, increasing the diameter to 10 m. The resulting limiting sensitivity of XEUS will be < 10<sup>-17</sup> ergs cm<sup>-2</sup>s<sup>-1</sup>, which is ~100 times greater than XMM and a factor of 10 greater than the AXAF-Chandra deep survey limit.

Parameter	Specification
Energy range	0.05 – 30 keV
Telescope focal length	50 m
Mirror effective collecting area @ 1 keV	5 m <sup>2</sup> (MSC1); >10 m <sup>2</sup> (MSC2)
Mirror effective collecting area @ 8 keV	$1 \text{ m}^2$ (MSC1); $1 \text{ m}^2$ (MSC2)
Angular resolution (HEW) @ 1 keV	2 arcsec
Field of view	5 - 10 arcmin

**Table 1.1:** XEUS mirror design. The Mirror Spacecraft (MSC) will be updated after several years of operation to increase the collecting area of the mirror modules [XEUS Science Case, 2000].

The "strawman" instrumentation package at launch includes a widefield imaging CCD camera (WFI) and two narrow field instruments. The WFI will have an energy resolution of 100 eV at 7 keV and cover a field of 5-10' diameter. The baseline design for the narrow field cryogenic imaging spectrometers (CIS) are an STJ array for the low energy band (0.5 - 1.5 keV) and a TES array for the high energy band (1 - 10 keV). The science that can be achieved with such detectors is shown in figure 1.1.



**Figure 1.1**: Estimated resolving power of the narrow field imaging spectrometers as a function of photon energy [XEUS Science Case, 2000]. The minimum resolving power necessary for diagnostics of various physical processes is also shown, illustrating how both the TES and STJ detector technologies could, in principle, achieve most of the required science.

Parameter	Specification
Energy resolution at 1 keV	2 eV
Energy resolution at 7 keV	5 eV
Field of view	30 – 100 arcsec
Count rate	250 Hz
Quantum efficiency	> 90% at 7 keV
Number of pixels in array	32 x 32 (minimum)
Pixel array packing efficiency	> 90 %
Pixel size	1 arcsec or 250 μm x 250 μm
Operating temperature	10 mK

**Table 1.2:** TES detector requirements for XEUS [Bavdaz, M., 1998]. This presumes the TES will be of the form of an imaging pixel array.

### **1.3 Laboratory Applications of TES Detectors**

In addition to the applications in astronomy described above, TES detectors have potential for use in ground-based X-ray microanalysis spectroscopy. X-ray microanalysis is a sensitive, non-destructive technique for providing spatially resolved chemical analysis, with many industrial applications. A sample is exposed to a finely focussed beam of X-rays or electrons (often from a scanning electron microscope) which excites characteristic X-rays. This fluorescent emission is measured with a spectrometer to deduce the chemical composition of the sample. In the semiconductor industry, two types of spectrometer are currently in use: semiconductor-based Energy Dispersive Spectrometers (EDS) and Wavelength Dispersive Spectrometers (WDS) [Hilton, G., 1999]. EDS systems dominate since they are inexpensive and offer rapid quantitative analysis. However, they are limited to energy resolution,  $\Delta E \sim 100 \text{ eV}$  which is insufficient to resolve overlapping peaks of interest such as the Si Ka and W Ma peaks in WSi<sub>2</sub> [Wollman, D., 2000]. Such peaks are resolved by WDS systems which use Bragg reflection from curved diffraction crystals and may have energy resolutions of 2 eV. The disadvantage with this method is the long time taken to serially scan the required energy range. Hence, neither EDS nor WDS systems fully meet the microanalysis requirements faced by a variety of industries. The semiconductor industry in the US has identified the need for improved X-ray detector technology to address this shortfall [NTRS, 1997]. This report highlights the inability of present techniques to provide fast and unambiguous analysis of small scale surface contamination (less than 0.3 µm in diameter) in semiconductors that has hindered the continued miniaturisation of integrated circuitry. Transition edge sensors are an attractive alternative since they combine the energy resolving power of a WDS with the speed of an EDS. Moreover, in contrast to a conventional EDS, the ability of a TES to resolve closely spaced X-ray lines at low energies means that it may be used in conjunction with low energy electron beams [Ladbury, R., 1998]. These are less penetrating, resulting

in superior analysis of surface contaminants and so a TES should be able to analyse particles as small as 0.08 µm [Wollman, D., 1997].

### **1.4 Thesis Structure**

Chapter 2 contains a description of the physics of phonon-mediated X-ray detection in transition edge sensors. Justification is given for our choice of absorber and thermometer materials. Modes of operation such as the use of electrothermal feedback are discussed and emphasis is placed upon sources of noise and methods of noise suppression. The equipment and cryogenic techniques used in the experiment are reviewed in Chapter 3. A new liquid helium dilution refrigerator was commissioned during this programme and a brief summary of the refrigerator and associated X-ray beamline are presented. A description is given of our prototype Superconducting QUantum Interference Device (SQUID) amplifier. Discussions of experimental techniques are continued in Chapter 4, in which testing of microchannel plates at low temperatures is described. In Chapter 5, the design constraints and fabrication methods for TES devices are explained. Results from investigations into TES reliability and materials science are presented in Chapter 6. This is an important field since degradation in the TES thermometer due to diffusion and alloy formation processes has proven to be a major obstacle to the development of stable devices. One potential solution to the reliability issue is to use iridium thermometers, and results from experiments designed to investigate the superconducting properties of iridium are discussed in Chapter 7. Chapter 8 includes the results of X-ray testing of a single pixel TES, with emphasis upon digital signal processing techniques. Astronomy applications demand imaging capability. One means of achieving this is to use extended absorbers with multiple TES output. This route is examined in Chapter 9 with particular emphasis on bismuth absorbers. Results of modelling heat flow through linear bismuth absorbers are presented. Finally, in Chapter 10, the results from this thesis are summarised and the aims of future research are outlined.

# **Chapter 2**

# The physics of x-ray detection with microcalorimeters

### 2.1 The Development of Microcalorimeters for X-ray Astronomy

A calorimeter may be used to determine the energy of an incident particle or photon by measurement of the resulting increase in temperature in the device. Calorimeter development can be traced back to the early days of nuclear physics and the radioactivity work of Pierre Curie. By the 1930s, small calorimeters had been operated at temperatures as low as 50 mK for the measurement of continuous fluxes, but it was not until 1975 that the first reference to the thermal detection of single particles or photons can be found [Niinikovski, T., 1975]. The first measurement of X-rays using a single photon calorimeter was made in 1984 by groups at the University of Wisconsin and NASA's Goddard Space Flight Center [McCammon D., 1984]. Since then, the development of microcalorimeters for X-ray instrumentation has been pursued by groups in the US, Europe and Japan. The present state of the art is currently defined by the National Institute for Standards and Technology (NIST, Boulder, Colorado). They have achieved an energy resolution of 2.0 eV FWHM at 1.5 keV with an Al/Ag Transition Edge Sensor [Wollman, D., 2000]. The Goddard group developed an array of 32 microcalorimeters with silicon thermistors and HgTe absorbers which was launched on the Japanese Astro-E satellite in February 2000 [Stahle C., 1997]. Unfortunately, Astro-E failed to achieve the correct orbit. In Europe, rapid progress has been made by the group at the Space Research Organisation of the Netherlands, SRON in Utrecht. They have a single pixel Ti/Au TES operating at 12 eV FWHM at 6 keV [Hoevers, H., 1999a] and are now working towards an imaging TES array.

Conventional X-ray detectors measure ionisation produced in the detector by photon absorption or by secondary interactions. For good energy resolution, the choice of materials is limited. For silicon the band gap energy is 1.2 eV but an average energy deposition of 3.6 eV is required to create an e-h pair [Fraser, G., 1989]. The excess energy (~ 70%) goes to produce phonons. The best resolution achieved with a Si-based X-ray detector (~ 120 eV FWHM at 6 keV) is an order of magnitude poorer than the TES results quoted above and is close to the predicted theoretical Fano limit. The Fano factor [Fano, U., 1946] describes the variance in number of electrons created in the initial, localised charge cloud and represents a fundamental statistical limit on energy resolution. Since most of the incident energy goes to produce phonons and the maximum energy of these phonons is only 60 meV, there are many more phonons than e-h pairs. Therefore, phonon detection is an inherently more sensitive technique offering better energy resolution for a given energy input. This process also allows low temperature devices to detect smaller energy depositions. Figure 2.1 shows comparison spectra from a

Si(Li) detector and a TES, illustrating the superior resolution of the TES. In addition, almost any material will generate phonons following X-ray absorption and so the choice of material is not so limited. This is important for experiments that require a particular nuclear species to be the absorber. For an X-ray detector, the absorber material may be a dielectric, normal metal or superconductor, depending on the required performance.



**Figure 2.1**: Example spectra of TiN from a conventional Si(Li) Energy Dispersive Spectrometer (EDS) and an Al/Ag bilayer TES [Hilton, G., 1999]. TiN is an important material in the semiconductor industry and such spectra indicate an important industrial application for TES technology. The superiority of the microcalorimeter to resolve spectral features is clear.

### **2.2 TES Microcalorimeter Description**

### **2.2.1 Thermal Detection of X-rays**

An X-ray microcalorimeter consists of an absorber in thermal contact with a thermometer (figure 2.2). The energy of the incident X-ray photon, E, is thermalised within the absorber and the resulting thermal distribution of phonons is measured by the thermometer. The increase in temperature,  $\Delta T$  provides a measure of the energy of the X-ray [Booth N., 1996]:

$$\Delta T = \frac{E}{C(T)} e^{\left(\frac{-t}{\tau_0}\right)} \qquad \text{where } \tau_0 = \frac{C(T)}{G(T)} \tag{2-1}$$

Where C(T) is the total heat capacity of the system (including the absorber, TES, bridge structure and connecting leads).

The intrinsic (without electrothermal feedback [Irwin, K., 1995a]) relaxation time of the microcalorimeter to reach equilibrium temperature has a 1/e time constant,  $\tau_0$  where G(T) is the thermal conductance between the heat bath and absorber. G(T) is defined by the thin (~1 µm) suspended Si<sub>3</sub>N<sub>4</sub> membrane structure on which the detector is fabricated. The role of the membrane is to weakly thermally couple the detector to the cold heat bath. Weak coupling to the heat bath is essential to enable the detection of thermal energy from the absorber within a few nanoseconds, improving thermalisation [Nahum, M., 1995]. The use of a thin membrane also reduces the probability of X-ray absorption events in the substrate. Some thermal coupling is required to cool the absorber after X-ray absorption and the thickness and span of the membrane determine the intrinsic time constant of a detector [Holmes, W., 1998]. Since  $\tau_0$  represents the pulse recovery time, it is an important factor in determining the maximum count rate of the device when operating in the absence of Joule heating. The effect of electrothermal feedback and Joule heating is described in section 2.5.

Typically, superconducting leads provide the electrical connection between the TES and wire bonding pads where connections are made to the readout SQUID (see Section 3.2) and bias circuitry. A superconductor with a transition temperature, Tc greater than the operating temperature of the device such as aluminium or niobium (1.2 K and 9.2 K, respectively) gives reliable electrical contact with negligible heat generation and excellent thermal isolation.



**Figure 2.2**: Schematic of a microcalorimeter incorporating a Transition Edge Sensor (TES) thermometer.

### 2.2.2 Types of Thermometer

Two types of thermometer are commonly used: semiconductor thermistors and superconducting transition edge sensors (TESs).

### **2.2.2.1 Semiconductor Thermometers**

Early development of X-ray microcalorimeters focused on highly doped semiconductor thermometers [Moseley, S., 1984 and McCammon, D., 1984], whose resistance is a strong function of temperature (varying as  $T^4 - T^{-9}$  in the temperature range 0.3 - 5 K [Coron, N., 1985]). These usually consist of a small (~1x10<sup>-9</sup> cm<sup>3</sup>) implant of Neutron Transmutation Doped (NTD) germanium, or ion implanted silicon. A dc voltage is applied to a series combination of the thermistor and a load resistor. Variations in thermistor voltage arising from X-ray absorption events are measured with a low-noise amplifier, typically a junction field-effect transistor (JFET) operating at 80 K. Optimisation of this configuration was given by Mather [1984] and impressive results have been obtained: 7.3 eV FWHM at 5.89 keV [McCammon et al, 1993]. Although this is 7 times worse than the fundamental thermodynamic energy resolution due to thermal fluctuations, it is comparable with contemporary results from superconducting TESs. However, improvements in the performance of semiconductor based devices require a reduction in the already very small heat capacity (0.1 pJ/K) which is an important factor in the design for astronomy devices where a relatively large heat capacity follows in

consequence from the need for useful quantum efficiency (which scales with absorber thickness). Constraints on the power dissipation in semiconductor thermistors limit the speed of response: for operation at 100 mK with optimum resolution the lower limit on the time constant is ~1 ms. Hence, superconducting TESs are currently favoured for future X-ray astronomy applications.

### **2.2.2.2 Superconducting TES Thermometers**

These consist of a thin (~ 100 nm) superconducting film biased on the transition from normal to superconducting states. A small perturbation in temperature, such as that due to absorption of an X-ray photon, results in a large change in resistance. Extremely high sensitivity may be achieved for narrow transition widths (~1mK), but the operating temperature for the detector must then remain stable to the order of 10  $\mu$ K [Wollman, D., 1997]. Another advantage to using superconducting thermometers include their lower impedance (R ~ 1  $\Omega$ ) compared to semiconductor thermistors (R ~ 10 M $\Omega$ ) [Stahle, C., 1995]. This means they are less susceptible to microphonic pickup and are also more readily impedance matched to Superconducting QUantum Interference Device (SQUID) amplifiers. When operated with electrothermal feedback [Irwin, K., 1995a], see section 2.5, superconducting thermometers enable the use of larger heat capacities (~10 pJ/K), which is important for astronomical applications, and can theoretically exceed the count rate limit of ~1 kHz of semiconductor thermistors [McCammon, 1993]. In this operating mode, a TES with a normal metal absorber has a lower fundamental resolution limit than any existing technology operating at the same temperature and absorber heat capacity and can tolerate higher count rates.

In general, the transition width of a superconductor scales with temperature, so the sharp transition required for microcalorimetry demands a low critical temperature superconductor. To achieve this, films made from pure metals with low transition temperatures such as tungsten (bulk  $Tc \sim 0.015$  K [Bruckmayer, M., 1997]) or iridium (bulk  $Tc \sim 0.112$  K) may be used. However, the transition temperature is highly dependent upon deposition conditions and reproducing films with the required Tc is often difficult. For example, thin films of tungsten usually deposit in the  $\beta$ -phase (an A15 structure), whereas bulk tungsten exists in the  $\alpha$ -phase (bcc), with transition temperatures as high as 4 K [Bond, W., 1965].

Alternatively, multilayers of normal metal and superconducting metal may be constructed with the transition temperature tuned to suit the application. The proximity effect [de Gennes, 1964] describes how a normal metal thin film in good thermal and electrical contact with a superconducting thin film will behave as a single superconductor with new properties. Addition of the normal metal reduces the transition temperature and the width of the transition. Such changes are dependent upon the relative thickness of the layers. For example, in the case of an Al/Ag (40 nm/80 nm) bilayer, the effect of the normal silver is to reduce the transition temperature of the aluminium from 1.2 K for pure Al to the

order of 110 mK [Finkbeiner, F., 1997]. A typical transition width is approximately 2 mK. An additional advantage to using a bilayer is the exploitation of the relatively long electron mean free path in normal metals such as Au, Cu and Ag, providing fast heat diffusion. A key concern with any multilayer system is reliability of the electrical properties of the TES. Interdiffusion of the materials across the interfaces may degrade the performance of the TES within hours of manufacture (Chapter 6). Single layer devices are less susceptible to degradation in this manner.

Many combinations of materials have been investigated with the aim of producing sharp transitions at ~100 mK that are stable over time. We have experimented with Al/Ag and Ti/Au bilayers, Al/Ag/Al trilayers, and single layers of iridium (see Chapters 5 and 6).

### 2.3 Energy Transfer: from Photon Absorption to Output Pulse

In a normal metal absorber at low temperatures, the energy of the incident photon is rapidly transferred into the electron system. When the X-ray is absorbed, a high energy inner-core electron is ejected which creates a local hot spot by electron-electron interaction. The time for thermalisation within the electronic system (the time to relax to a Fermi-Dirac distribution) has been shown experimentally to be of the order of nanoseconds; significantly faster than computational predictions [Pothier, H., 1997]. The electron system is coupled to the phonons and a thermal distribution of phonons is established which diffuses into the superconducting thermometer. These relatively high energy phonons break the Cooper pairs (bound electrons) in the superconductor releasing quasiparticles and more phonons. The Cooper pair binding energy (0.34 meV for Al) is extremely small compared to the band gap of insulators and semiconductors (1.2 eV for Si), and is also small compared to the Debye and Fermi energies (36.9 meV and 11.7 eV, respectively for Al). The Debye energy can be considered to be an approximation to the maximum phonon energy, and similarly, the Fermi energy is approximately the maximum energy of electrons in the electron 'gas' of a metal. Therefore, thermal phonons from the absorber (and electrons if the absorber is in electrical contact with the TES) readily break Cooper pairs in the superconducting thermometer into quasi-electrons and quasi-holes, collectively quasiparticles. The increase in quasiparticle population represents the transition from superconducting to the normal state. This rapidly increases the resistance of the superconductor and the resistance change,  $\Delta R$  is measured to yield the temperature change resulting from the absorption of the X-ray photon. Assuming a linear detector response, the temperature rise is proportional to the energy of the incident X-ray.  $\Delta R$  is measured using a SQUID amplifier (Superconducting QUantum Interference Device), see Chapter 3 for a SOUID description. A change in TES resistance leads to a change in the current to the input coil of the SQUID, as can be seen in figure 2.3. The SQUID response is easily measured with room temperature electronics.



**Figure 2.3**: Bias circuit (top) and thermal circuit (bottom) of the TES [Roth, P., 2000a]. The increase in TES temperature following an X-ray event increases the resistance of the TES. From the bias circuit, it is clear that the resulting change in the resistance ratio of the shunt resistor and TES reduces the current to the SQUID input coil. The thermal circuit follows the nomenclature of Roth [2000a] illustrating the power flow between the absorber (subscript a), TES (subscript s) and cold heat bath (subscript b). In this simple model, the energy from a single photon is assumed to be completely absorbed in the absorber. No distinction is made between phonon and electron temperatures in the absorber,  $E\delta(t)$  flows through the thermal conductance  $G_{as}$ , since  $G_{as} >> G_{ab}$ .

### 2.4 Absorber Material

The constraints upon the absorber include the need to efficiently thermalise the incident X-ray energy, low heat capacity to minimise thermodynamic fluctuations, and high quantum efficiency (QE). High

QE comes from using thick absorbers or materials with high Z (since the photoelectric cross section is proportional to  $Z^4/E^3$  [Bragg, W., 1914]):

$$QE = 1 - e^{(-\mu d)}$$
(2-2)

Where  $\mu$  is the linear absorption coefficient of the absorber medium and d is the absorber thickness.

Heat capacity is minimised by restricting the volume of the absorber within the limits imposed by QE and the required count rate for a given input X-ray flux. Efficiency of thermalisation (or the rate at which the excited phonon distribution relaxes to a thermal distribution) dictates that the material should be metallic and ideally a normal metal rather than a superconductor. The merits of each type of absorber are discussed below.

The fundamental difference between dielectric and superconducting absorbers is the level of the Debye energy, which corresponds approximately to the maximum phonon energy. In dielectrics it is always less than the band gap, but for superconductors the Debye energy exceeds the energy gap [Booth, N., 1996].

Insulators, semiconductors and superconductors do not make ideal absorbers. These materials have phonon spectra which relax slowly and inefficiently to thermal distributions. For insulators and semiconductors the problem lies in charge trapping. Such materials have impurity levels in their bandgaps on which the electrons can be trapped before thermalising giving noisy and incomplete thermalisation. In addition, there are other decay processes which are not fully understood such as anharmonic decay (arising from inelastic deformation of the atomic lattice) and isotope scattering [Blakemore, J., 1974]. These are further complicated by crystal anisotropy leading to phonon focusing, differences between transverse and longitudinal modes of propagation and the fact that the group velocity increases as phonon energy decreases [Booth, N., 1996]. Such 'quasi-diffusive' transport was first observed in  $\alpha$ -particle absorption in silicon crystals [Peterreins, T., 1988].

Dielectric absorbers for detectors of athermal phonons can be improved by the use of quasiparticle trapping. A large fraction of the surface area of the absorber is covered with a thin film of a high gap superconductor (aluminum pads) in electrical contact with a low gap superconductor (tungsten) thermometer. The incident X-ray produces a mean athermal phonon energy of  $\sim 2 \text{ meV}$  in the absorber. This is significantly larger than the Cooper pair binding energy of aluminium ( $\sim 0.3 \text{ meV}$ ) so the phonons readily produce quasiparticles in the Al film. These diffuse into the tungsten via the electrical contact in a characteristic time much faster than the recombination lifetime and are trapped there by the energy gap difference. By definition, the tungsten will have many thermally excited quasiparticles and so the transfer of energy into the tungsten is primarily by quasiparticle-quasiparticle interaction which is a rapid thermalisation process. If the tungsten thermometer was purely

superconducting then the quasiparticles incident on the TES would create phonons, slowing the thermalisation process. The device can be operated in ETF mode (section 2.5) and a thin film of Al is used so that the heat capacity of the system is not adversely affected. Early results from the Stanford group working with 1 cm x 1 cm x 1 mm Si absorber are encouraging (FWHM < 350 eV at 6 keV), but so far at least 40 % of the energy is lost to subgap phonons escaping into the substrate when the quasi particles are initially created in the high gap superconductor [Irwin, K., 1995c]. A more direct means of exploiting the rapid transfer of energy through the electron system is to use a normal metal absorber, and this is our approach.

In superconductor absorbers most of the deposited energy excites the phonon spectrum which relaxes to a thermal distribution in a slow and complicated manner. A group in Genoa have found that below 100 mK thermalisation is not complete on ms timescales for superconductors with high Debye temperatures, although it is for those with low Debye temperatures [Cosulich, E., 1993]. Energetic phonons (phonon energy > superconducting bandgap) have a very short mean free path for breaking Cooper pairs which recombine by releasing another phonon which is free to break further Cooper pairs. Thermal equilibrium is reached when the rate at which Cooper pairs are broken is balanced by the rate of quasiparticle recombination, creating Cooper pairs and emitting a low energy phonon. Phonons with energies less than the Cooper pair binding energy thermalise by scattering off quasiparticles. Quasiparticle excitations in a superconductor far below the critical temperature where the ambient quasiparticle population is low, quasiparticle recombination times are slow. Therefore, some of the deposited energy thermalises very slowly and registers in the thermalisation occurs under the

condition  $\frac{T}{\theta} < 3 \ge 10^{-4}$  where T is the operating temperature and  $\theta$  is the Debye temperature [Cosulich, E., 1993]. The future of superconducting absorbers looks set to include incorporation of small normal metal regions to stimulate quasiparticle recombination with a limited impact on heat capacity. Another less promising approach is to trap magnetic flux in a superconducting film to improve thermalisation [Stahle, C., 1994].

In contrast, in a normal metal absorber the deposited energy is almost entirely thermalised by electron excitation which relaxes rapidly and efficiently to a thermal distribution allowing faster detector response and improved energy resolution [Martinis, J., 1997]. A normal metal absorber is free of metastable states and so has no long-lived athermal excitations [McCammon, D., 1993]. Such excitations have an associated statistical variation and thus a degrading effect on energy resolution. The electrons in the absorber cool by emitting phonons that propagate into the superconductor. Therefore, electron-phonon coupling in the absorber is an important process in the transfer of heat from the X-ray event into the thermometer.

In summary, the ideal absorber must be a compromise between high Z (for stopping power of X-rays) and low heat capacity, C to reduce the thermodynamic noise which degrades energy resolution. It is desirable for most of the absorbed energy to go directly into the electronic system and this requires a material with an electronic component to the heat capacity dominant over the lattice component. At 100 mK, the semi-metal bismuth (Z = 83) meets these criteria. Bismuth has a remarkably small electronic heat capacity resulting in a heat capacity ~ 40 times lower than that of gold (which corresponds to an improvement in intrinsic energy resolution of  $\sim 6$  times). This is a consequence of the low free carrier density of ~  $3 \times 10^{17}$  cm<sup>-3</sup>, equivalent to  $10^{-5}$  per atom [Pratt, W., 1978]. The low heat capacity and poor thermal conductivity of bismuth are shared by other elements of the Pnictogen group of the Periodic table, such as antimony (Z = 51) and arsenic (Z = 33). These are also semimetallic (metalloid) solids but have electronic coefficients of the heat capacity at least 5 times larger than that of bismuth (0.112 mJmol<sup>-1</sup>K<sup>-2</sup> for antimony compared to 0.021 mJmol<sup>-1</sup>K<sup>-2</sup> for bismuth [American Institute of Physics Handbook, 1972]). Hence, bismuth is the preferred absorber material. It should be noted, however, that there is a limit to the possible reduction in heat capacity. The absorber material should be chosen so that the maximum expected input energy drives the thermometer no more than about one tenth of a transition width [Irwin, K., 1995a]. Larger digressions from the equilibrium position result in lengthened pulse duration and distortion of the pulse tops which complicates pulse analysis. If low heat capacity materials such as bismuth are used in the absorber, incident energy from X-rays is often sufficient to send small absorbers normal. Composite absorbers utilising a layer of bismuth over gold or copper have been proposed but, as Irwin noted [1995a], perhaps the most important use of bismuth will be for large area absorbers (square cm) for which bismuth is possibly the only material that could sustain a few eV resolution. This geometry, however, places inherent limits on the possible speed and count rate due to the relatively low thermal conductivity of bismuth (the phonon thermal conductivity reaches a maximum of ~ 86 W cm<sup>-1</sup>K<sup>-1</sup> at a temperature of 3.5 K [Issi, J-P., 1977], and scales as  $T^3$  for T < 3.5 K). A full discussion of bismuth absorbers is given in Chapter 9.

### 2.5 Electrothermal Feedback (ETF)

Negative electrothermal feedback [Irwin, K., 1995a] is a method used to simplify TES operation and yield faster response times allowing higher count rates. The superconductor film is voltage biased and the current flowing through the film (less than the critical current) is measured using the SQUID. The heat bath is cooled to well below the transition temperature of the superconductor. As the film cools by loss to the heat bath, its resistance drops and so Joule heating  $(V^2/R)$  increases. The film will self-regulate in temperature at the point where Joule heating from the voltage bias equals the heat loss to the bath via the substrate. The bias conditions can be adjusted so that this occurs on the superconducting - normal transition.

When an X-ray photon interacts with the absorber, the temperature and hence resistance of the film rises. This decreases the measured current and so Joule heating is reduced, but the heat loss to the bath remains approximately constant (since the change in temperature is typically very small, < 0.5 mK). The energy of the x-ray is removed by a reduction in Joule heating which gives a faster thermal response time,  $\tau_{eff}$  than the intrinsic thermal relaxation time,  $\tau_0$  [Booth, N., 1996]:

$$\tau_0 = \frac{C}{G} \qquad \qquad \tau_{eff} = \frac{\tau_0}{\left(1 + \left(\frac{\alpha}{n}\right)\right)} \tag{2-3}$$

Where  $\alpha$  is a dimensionless measure of the sensitivity of the TES and *n* depends on the dominant thermal impedance between the electrons in the TES and the substrate.

 $\alpha$  is derived from the logarithmic gradient of the superconducting transition. It is defined as:

$$\alpha = \frac{d(\log R)}{d(\log T)} = \left(\frac{T}{R}\right) \left(\frac{dR}{dT}\right)$$
(2-4)

For 
$$\alpha = 40$$
 and  $n = 5$ ,  $\tau_{eff} \sim \frac{\tau_0}{10}$ 

Since the gradient of the transition varies with temperature, quoting a single value for  $\alpha$  can be misrepresentative (see figure 2.4). In the literature,  $\alpha$  values of ~ 1000 are often quoted [Irwin, K., 1992] and this is possible as *R* tends to zero, but for stability under bias conditions it is more usual to operate at ~ 10 % of the normal resistance value where  $\alpha$  values of 40 - 50 are typical. From this simple analysis, it would appear that a high  $\alpha$  value, and hence steep transition, is advantageous. Recent results show, however, that a large  $\alpha$  is associated with an excess noise component which acts to degrade instrument resolution [Hoevers, H., 1999a]. Hoevers et al. report that the total current noise of their TES includes a high frequency component which appears to reflect trends of the  $\alpha$  curve, attributed to thermal fluctuation noise within the TES.

Repeated measurements of transitions are required to ascertain whether structure in the transition is real or a consequence of errors in temperature measurement, which are difficult to quantify. Figure 2.4 shows how structure in the transition can affect the computed values for alpha.



**Figure 2.4**: The variation of  $\alpha$  for a Au/Ti TES transition, measured in our laboratory. All transitions in this thesis are subject to similar uncertainties regarding apparent fine structure, which may result from measurement errors.

The parameter *n* is an important factor in the energy flow through the system. In general, the heat flow from the TES at a temperature  $T_c$  into the Si<sub>3</sub>N<sub>4</sub> substrate at a temperature  $T_{SUB}$  is proportional to  $(T_c^n - T_{SUB}^n)$ . For thicker films at higher temperatures, Kapitza boundary resistance (enhanced thermal resistance across a boundary at liquid helium temperatures [Kapitza, P., 1941]) is dominant and n = 4. For thinner films at lower temperatures, electron - phonon decoupling in the film dominates and n = 5 or 6, depending on temperature [Mels, W., 1998]. In thin films the Kapitza boundary resistance will be small since the phonon distribution is no longer separate from that in the substrate. Therefore, in our case of thin films at low temperatures (T < 200 mK), the most important process in the transfer of heat from the electron system to the substrate is the weak electron - phonon coupling, and so  $n \sim 5$ .

The energy of the event is determined by finding the integral of the change in Joule power. This simplifies pulse analysis and the energy can be measured in the absence of noise with no free parameters. The inclusion of low frequency 1/f noise will complicate this analysis, although in our experiments we observed no measurable 1/f component of noise (see Chapter 8). Non-linearities in the resistive transition and physical non-uniformities in the bilayer will effect pulse shape, but not the pulse integral and so do not degrade energy resolution directly. Even events which saturate and drive the film normal can be measured because the energy loss is small as long as the pulse duration is small compared to  $\tau_0$ . The energy collection efficiency can be measured directly: the product of the bias voltage and the integral of a current pulse is divided by the known incident energy value. A result of unity would imply perfect energy collection. Most of the energy loss is through thermal leakage to the substrate. This thermal path is necessary for electrothermal feedback, but is always minimised to reduce energy loss from the X-ray. A typical value for *G* is 1 nW/K.

### 2.6 Factors Affecting Energy Resolution

An intrinsic baseline energy resolution may be defined which includes the detector response and noise from the SQUID electronics. Added to this in quadrature are the many independent components of the pulse processing chain to give the practical FWHM of the device. These secondary sources of resolution degradation typically add several eV to the FWHM at 6 keV, and are particularly significant if non-optimal shaping of the output pulse or multichannel analysers with relatively few channels are used (for example, 4096 channels operating over 10 keV gives binwidths of ~3 eV, placing a lower limit on resolution).

### 2.6.1 The Thermodynamic Limit

The baseline energy resolution of a TES spectrometer operating without ETF is limited by thermodynamic fluctuations in the absorber which are determined by the heat capacity.

The heat capacity C(T) for any material is defined as the sum of the lattice and electronic components [Walton, A., 1983]:

$$C = \left(\frac{125T}{\Theta}\right)^3 + T\gamma$$
 mJ/mole/K where  $\Theta$  is the Debye temperature (2-5)

For normal metals at low temperatures the linear electronic term,  $\gamma$  dominates.

For superconductors the electronic contribution is proportional to  $e^{\left(\frac{-T_c}{T}\right)}$  and so is negligible for  $T \ll T_c$ . Thus, for superconductors well below their transition temperatures and for pure dielectric crystals, C(T) has the dependence:

$$C(T) = 1944 \left(\frac{m}{M}\right) \left(\frac{T}{\Theta}\right)^3 \qquad J/K \qquad (2-6)$$

Where m is the mass of the absorber and M is the molecular weight of the absorber.

To estimate the thermodynamic limit on energy resolution, consider the phonon system in the detector. The mean number of phonon modes, N is  $\frac{C(T)}{K_B}$ , and typically each mode has a quantum occupation number 1 and rms fluctuation of 1 phonon. The mean energy per mode is  $K_BT$ , so the mean square energy fluctuation is  $N(K_BT)^2 = K_BT^2C(T)$ . Therefore, the rms. energy fluctuation is:

$$\boldsymbol{\sigma} = \zeta \left( K_B T^2 C(T) \right)^{1/2} \tag{2-7}$$

where  $\zeta$  is typically 1.5 - 2.0 (without ETF). Note that  $\sigma$  is not a function of E.

The same expression can be determined (less intuitively) from the derivatives of the partition function, regardless of the details of the system [Paerels, F., 1997].

These fluctuations occur on the timescale of the thermal time constant,  $\tau_0 = \frac{C}{G}$ . The accuracy of measurement of energy deposited in the detector on shorter timescales can exceed the thermal rms limit of  $\Delta E_{rms} = \sqrt{K_B T^2 C(T)}$ , if frequencies higher than  $\tau_0$  are considered. Without ETF, this is not practical because Johnson noise typically dominates the signal for high frequencies. However, with ETF the  $\zeta$  factor can be reduced by an order of magnitude:

$$\varsigma = 2 \left(\frac{n}{2}\right)^{1/4} \left(\frac{1}{\alpha}\right)^{1/2}$$
 [Booth, N., 1996] (2-8)

$$\sigma = 2^{3/4} \left( K_B T^2 C(T) \right)^{1/2} \left( \frac{\sqrt{n}}{\alpha} \right)^{1/2} \text{eV}$$
 (2-9)

$$\Delta E_{FWHM} = 2.36 \Big( K_B T^2 C(T) \Big)^{1/2} \left( \frac{\sqrt{8n}}{\alpha} \right)^{1/2} \text{eV}$$
 (2-10)

From equation (2-10), ETF reduces energy resolution over the non-ETF thermodynamic limit by a factor of ~  $2/\sqrt{\alpha}$ , hence the desire for large  $\alpha$  values. If an optimum filter is applied to the exponential pulse in the presence of Johnson and phonon noise then we get equation (2-10). Note that although C is a function of T, C can be treated as a constant since temperature changes are small (~ mK).

As a numerical example, we now calculate the energy resolution limit for gold and silver absorbers.

For current XEUS resolution and pointing accuracy specification (table 1.1) we require the minimum side length of a square, single pixel absorber, to be  $L_{min} = 250 \,\mu\text{m}$ .

Assuming a QE of 90% at 6 keV, an absorber thickness  $d > 2.9 \ \mu m$  is required (from equation 2-2). The mass of gold absorber, *m* is therefore:

$$m = \rho \ge 2.9 \ge 250 \ge 250 \ \mu m = 3.5 \ge 10^{-6} g$$

Bulk density,  $\rho$  of Au = 19281 kgm<sup>-3</sup> [bulk density,  $\rho$  of Ag = 10500 kgm<sup>-3</sup>]

From equation (2-5), specific heat of gold, C = 0.0754 mJ/mole/K

Atomic mass of gold A = 197 g/mole [for silver A = 108 g/mole] so the gold absorber contains 1.77 x 10<sup>-8</sup> moles.

 $\therefore C = 1.34 \text{ x } 10^{-12} \text{ J/K}$ 

 $\therefore \Delta E = 2.5 \text{ eV} \text{ [from (2.10) with } n = 5, \alpha = 40, T = 100 \text{ mK]}$ 

Alternatively, for an energy resolution of < 2 eV the corresponding limit on d is:  $d < 2.0 \,\mu\text{m}$ , or QE < 80 % (as shown in figure 2.5).

For comparison, at 6 keV a 2  $\mu$ m silver absorber is 62 % efficient. This is adequate for many astronomical applications but these simple calculations show there is a clear trade off between QE (thickness, d) and energy resolution (heat capacity, C).

From this simple estimate, the 2 eV resolution and 90 % QE specified for can only be met by increasing the performance enhancement of ETF. To do this, the transition temperature must be reduced or the  $\alpha$  value increased. Attempts to use high  $\alpha$  values have induced excess noise and lower operating temperatures can lead to stability problems [Hoevers, H., 1999a]. Generating the required energy resolution and quantum efficiency in one device remains one of the key outstanding issues for a space instrument.



Figure 2.5: Trade-off curves between QE and energy resolution assuming n = 5,  $\alpha = 40$  and T = 100 mK and a 250 x 250  $\mu$ m<sup>2</sup> metal absorber.

### **2.7 TES Noise Characteristics**

The noise in a TES detector is due to the combination of a variety of uncorrelated sources: phonon noise in the link to the heat bath, Johnson noise in the resistor and an observed excess noise component. In addition to this there is electrical noise input from the bias and readout circuitry, but this will not be discussed here. In recent experiments, the excess noise component has been unexpectedly large and is perhaps the most interesting theoretically. Consideration of this noise has redefined some of the design parameters for a TES, such as its normal state resistance and thermal conductance. Each of the noise sources are discussed below.

### 2.7.1 Phonon Noise

Phonon noise arises from spontaneous energy flowing through the device. This is a continuous process produced by random propagation of phonons (or electrons or quasiparticles) in the heat link between the device and the cold bath. Phonon noise dominates over Johnson noise at low frequencies, particularly for large  $\alpha$ . For higher frequencies the white Johnson noise spectrum dominates. Assuming non-ETF operation, the phonon noise spectrum is flat below a knee at  $\frac{1}{\tau_0}$  where  $\tau_0 = \frac{C}{G}$ 

and falls off above this (figure 2.6) [McCammon, D., 1993]. ETF does not modify the shape of the spectrum, only the position of the knee and the dependence of the phonon and signal noise power densities on  $\alpha$ . Phonon noise follows the same frequency dependence as the output signal and so the measurement error from this noise can be reduced by choosing a sufficiently large bandwidth. However, there are limits on how large the bandwidth can be. If only frequencies below the knee are used then it is not possible (without ETF) to exceed the thermodynamic energy resolution limit,  $\Delta E_{rms} = \sqrt{K_B T^2 C(T)}$ . For this, use must be made of frequencies faster than the knee. Increasing  $\alpha$ lifts the phonon noise and signal above the Johnson white noise (scaling as  $\alpha^2$ ), linearly increasing the point at which the signal drops below the Johnson noise, expanding the useful bandwidth. This is only of benefit up to the thermalisation frequency. Thermalisation time places an upper limit on the frequency space, since no information exists faster than the time required to thermalise energy in the absorber. Therefore, it is desirable to lower the knee and hence to use detectors with long intrinsic time constants, and hence weak thermal conductance, G. ETF enables these constraints to be met without compromising the effective count rate of the device by keeping the intrinsic time constant slow yet utilising a much faster effective time constant. The knee position is at a higher frequency with ETF due to the use of the effective time constant, but below this frequency the phonon and signal noise power spectral densities show no dependence on  $\alpha$ . This means that under ETF with large  $\alpha$ , energy resolution can exceed that of the thermodynamic limit without cause to extend the bandwidth beyond the knee.





Frequency (arbitrary log scale)

Figure 2.6: Signal-to-noise for an ideal calorimeter [McCammon, D., 1993]

### 2.7.2 Johnson Noise

Within the TES resistor, random electron density fluctuations cause changes in the associated electric field giving rise to Johnson noise. An important point made by Mather [1982] is that Johnson noise is modified under ETF. A weak thermal link to the heat bath is required for the operation of a TES but this means that the bias current dissipates power in the Johnson noise source. By the nature of negative feedback, an increase in the Johnson noise will increase the temperature of the resistor, increase the resistance and thereby reduce the bias current and current noise at low frequencies. In other words, the bias voltage does work on the current noise in the TES by producing temperature fluctuations which oppose the original noise source. Mather claims a 60% reduction in Johnson noise due to ETF and Irwin noted [1995b] that for extreme negative feedback the Johnson noise component is negligible for frequencies below the knee in the signal spectrum ( $\omega^2 \tau_{eff}^2 \ll 1$ ). For higher

frequencies, it reverts to a white noise spectrum,  $I_J = \sqrt{\frac{4K_B T_0}{R_0}}$ . This is because the electrothermal

feedback has a minimum response time approximately equal to the effective time constant.

### 2.7.3 Contact Shot Noise

Shot noise from contact between the resistor and the bias rails can exceed Johnson noise, but only in detectors with high (~  $1M\Omega$ ) dynamic impedance such as germanium bolometers, and not in low impedance TES devices [Mather, J., 1982].
The measured energy resolution of X-ray TES microcalorimeters is often larger than predictions based on the fundamental thermodynamic limit. For example, Hoevers, et al. reported a measured resolution of 60 eV at 5.89 keV with a detector with a thermodynamic limit of 23 eV [Hoevers, H., 1999b]. This discrepancy was attributed to an observed excess current noise in the TES. The measured current noise typically exceeds that expected from the sum of SQUID current noise, Johnson noise and phonon noise, for all bias voltages. The source of this excess component has been the subject of much recent discussion. Proposals include the movement of flux vortices in the TES [van Gurp, G., 1968, 1969] and 1/f noise arising from thermodynamical temperature fluctuations [Gildemeister, J., 1999]. Each of these are reviewed briefly in sections 2.7.5 and 2.7.6, but the source of the excess noise remains unresolved. In general, low  $\alpha$  values appear to be favourable to good energy resolution, to the detriment of attainable count rate.

# 2.7.5 Flux flow

The physics of the transition to superconductivity is not clearly understood for current biassed thin film devices. Irwin et al. [1998] conclude that the resistive state for a TES superconductor with a long electronic mean free path arises from the formation of phase slip centres. These are small dissipative regions (of size comparable to the coherence length) with resistance significantly less than the normal state resistance, where the order parameter varies periodically. However, other authors [Stuivinga, M., 1983] note that phase slip centres are only energetically favourable under certain geometric conditions. They have been observed in narrow one-dimensional wires but in wider films it has been necessary to cut the sample to form a constriction. Stuivinga et al. report that even long notches, of length comparable to the order parameter,  $\psi$  (where  $|\psi|^2$  can be treated as the density of superconducting electrons) and width equal to half of the strip width, were only able to weakly constrain the position of the core of the phase slip centre. Therefore, the occurrence of phase slip centres in relatively wide, uniform films may not be energetically favourable. Steps and hysteresis in the current-voltage curves are associated with phase slip centres [Skocpol, W., 1974] and these have been observed by Wollman in the operation of their detectors [Wollman, D., 1997]. These features can be removed by the application of a small magnetic field. We have not seen any step features in our I-V curves.

An alternative explanation derives from the well known fact that Type I superconducting thin films show Type II behaviour in the presence of transverse magnetic fields [Tinkham, M., 1963]. For a Type I superconductor, as the external transverse magnetic field is increased there is some flux penetration into the surface creating laminar regions of flux. In contrast, a similar magnetic field will penetrate a Type II superconductor in isolated vortices of singly quantised flux. Even if our TES is of Type I, the bias current will create transverse fields at the edges of the film. The vortices generated are multiply

quantised (as opposed to singly quantised for true Type II superconductors) and migrate across the film perpendicular to the current direction. Interaction between the flux tubes and the bias current causes flux flow to occur and gives rise to the measured resistance. As the bias current is increased, dissipation in the core of the flux lines increases driving regions of the film normal. This continues until all of the film is in the normal state. Evidence for this resistance mechanism comes from the observations of resistive state generation in current biased films under zero applied field. Flux flow is known to be a source of voltage fluctuations, and current noise. However, there is strong evidence from a number of sources [Hoevers, H., 1999a; Clarke, J., 1976] to suggest that in the case of an X-ray bolometer, thermal fluctuations are a more likely source of the observed excess noise.

#### 2.7.6 Thermal Fluctuations

The thermal fluctuation model of Voss and Clarke [1976] has been widely used to interpret excess noise seen at the superconducting transition of thin films. Their model describes how resistance fluctuations arise from equilibrium thermal fluctuations. It is intuitively useful to define a fluctuating temperature through the relationship  $\Delta E = C_v \Delta T$ , but strictly speaking this is model of spontaneous enthalpy fluctuations. For voltage fluctuations in the presence of a current:

$$<\Delta V^{2} >= V_{DC}^{2} \beta^{2} K_{B} T^{2} C_{V}^{-1}$$
(2-11)

where 
$$\beta = \left(\frac{1}{R}\right) \frac{dR}{dT}$$
 and  $\Delta V_{DC}$  = the average value for the voltage

The frequency spectrum is given by using the Langevin diffusion equation and so does not give any explicit 1/f region. A number of authors [Voss, R., 1976; Richardson, J., 1950; Lundstrom, L., 1973] generated 1/f regions by postulating the existence of anomalous behaviour at long wavelengths in the mean square amplitude for oscillations or in the coupling constant of the voltage to the fluctuating quantity. However, such additions to the model are hard to rationalise. This is unfortunate since the general form of the model is a good fit to many experiments. The model is most successful in situations where  $\frac{dR}{dT}$  is large, strongly coupling thermal fluctuations to voltage fluctuations, ensuring that thermal noise sources dominate. The superconducting transition is a good example of these conditions. Clarke and Hsiang [1976] investigated the transitions of tin and lead films and concluded that the noise scaled inversely with sample volume (or specific heat) and was proportional to  $(TR)^2$ 

 $\left(\frac{dR}{dT}\right)^2$  and  $V_{DC}^2$ , all in good agreement with equation (2-11). Whether the noise spectrum shows a 1/f

region seems highly dependent upon geometry, and in particular the thermal coupling of the sample to the substrate. Freely suspended tin films show clear 1/f regions [Ketchen, M., 1978] whereas tin and

lead films mounted on a substrate with an aluminium interface layer show a markedly flattened spectrum at low frequencies (<30 Hz) [Clarke, J., 1976]. In the context of TES's, our collaborators at SRON have shown [Hoevers, H., 1999a] that their excess noise is well described by the Voss and Clarke model. It can be easily distinguished from flux flow noise since the latter shows no explicit dependence on  $\alpha$ . Clarke and Hsiang also dismiss flux flow noise as a possible source of the excess noise on the grounds of a discrepancy of 5 orders of magnitude at zero frequency, the dependence of

the noise on  $\left(\frac{dR}{dT}\right)^2$  and the reduction in noise (as opposed to an expected increase for flux flow)

when an external magnetic field is increased.

# **Chapter 3**

# Equipment and experimental techniques

In this chapter, details of the equipment and techniques used to investigate detectors at low temperature are presented.

### 3.1 The Liquid Helium Dilution Refrigerator

All of the ultra low temperature experiments conducted as part of this thesis used an Oxford Instruments KelvinOx 100 liquid helium dilution refrigerator with a cooling power specification of 100  $\mu$ W at 100 mK. Dilution refrigerators have become a standard laboratory tool for the production of ultra low temperatures for many different research purposes.

#### **3.1.1 Principle of Operation**

Cooling to a base temperature of 10 mK is achieved by exploitation of the fact that liquid helium can exist in two stable isotopes: <sup>3</sup>He and <sup>4</sup>He. When a mixture of these two isotopes is cooled below a tricritical point at 860 mK, a gravity driven separation into two phases occurs. The tri-critical point is defined as the temperature at which a helium mixture may exist in the normal state, superfluid phase or begin to separate into 'concentrated' and 'dilute' phases. The concentrated phase is rich in <sup>3</sup>He and is less dense than the dilute phase which is rich in <sup>4</sup>He. The enthalpy of the <sup>3</sup>He in the two phases is different and so it is possible to obtain a cooling effect by 'evaporating' the <sup>3</sup>He from the concentrated phase into the dilute phase [Wilks, J., 1970]. Although a full analysis of the liquids requires a quantum mechanical description, the effect is analogous to cooling by evaporation from a liquid surface. The phase boundary is set up in the mixing chamber, onto which we mount our experiment (figure 3.1). Cooling from this process is sufficient to reduce the temperature from 860 mK to 10 mK. Temperature control is by means of a heater on the mixing chamber, which is set to work against the cooling power until the correct temperature is achieved.



**Figure 3.1**: Simple experiment mounting scheme for X-ray testing. The TES detector and SQUID are mounted on an oxygen-free (for high thermal conductivity at low temperature) copper bracket that hangs from the mixing chamber. The SQUID is encased in a niobium canister for magnetic shielding.

# **3.1.2 Technical Description**

Our refrigerator was custom made by Oxford Instruments Superconductivity Ltd. (Tubney Woods, Abingdon, Oxon, UK) for the development of X-ray detectors and includes side access to the experiment chamber (figures 3.2 and 3.3). This was designed to permit the use of external X-ray sources which can be readily adjusted for count rate and energy, or switched off completely for noise measurements (section 3.1.3). When operational, it will be the only facility of this type in the UK available for X-ray detector development. The refrigerator also has extra ports to the inner vacuum chamber (IVC) so that additional wiring can be added to accommodate future experiments. Currently, the experiment wiring loom to the 1 K plate consists of 6 manganin twisted pairs and 4 copper twisted pairs. Below the 1 K plate the copper-coated niobium-titanium wiring is superconducting during operation. All wiring is thermally anchored at the 77 K and 4 K levels to reduce the heat load to the mixing chamber.



**Figure 3.2**: Liquid helium dilution refrigerator mounted in an A-frame vibration isolation support structure. The sidearm for use of external X-ray sources is shown.

#### **3.1.3 Beamline**

For future X-ray testing, we will couple a soft X-ray source beamline onto the sidearm, capable of supplying X-rays from 200 eV – 5 keV. This X-ray source is a heated cathode with copper anode. Various coatings may be applied to the anode to generate fluorescent X-rays of the required energy within this range. For example, a carbon coating (SiC) generates  $K_{\alpha}$  line X-rays at 277 eV and 1740 eV and magnesium (MgO) gives 525 eV and 1.253 keV. The maximum voltage that can be applied is 5 kV.

This system will be the only facility in the UK available to characterise the performance of microcalorimeter X-ray detectors over their full energy range. All other facilities use radioactive sources mounted in the refrigerator, which is also our approach until the beamline is operational. However, the sidearm currently has the disadvantage of complicating access to the mixing chamber. Both the 4 K and 77 K thermal shields extend to the end of the sidearm to reduce the solid angle of thermal radiation load onto the mixing chamber (figure 3.3). This means that it is not possible to extract the insert from the top of the refrigerator as in conventional 'top-loading' designs and so we must disassemble the lower section of the refrigerator to gain access to the experiment. This adds

considerably to the turn-around time of the refrigerator cooldown cycle (approximately 2 weeks) and also increases the risk of introducing vacuum leaks in the system. To reduce this risk, an alternative mounting arrangement to that shown in figure 3.1 has been fabricated which enables us to attach the experiment to the mixing chamber by breaking fewer vacuum seals. This is shown in figure 3.3 and incorporates a single-point location to the mixing chamber and electrical connectors designed to be attached/removed by use of a purpose-built tool. With these features, it is possible to locate the experiment on the mixing chamber using the limited access provided by breaking the lowermost vacuum seals only, leaving the sidearm intact.



**Figure 3.3**: Coss-section of experiment chamber of the refrigerator showing the sidearm, inner and outer vacuum chambers (IVC/OVC), 4 K and 77 K thermal shields and the TES/SQUID mounting arrangement to provide X-ray beamline axis alignment of the TES [Favill, B., 2000].

#### 3.1.4 Vacuum System

We have experimented with both oil diffusion pumps and turbo-molecular pumps for maintaining the vacuums in the outer and inner vacuum chambers of the refrigerator (figure 3.3). Oil diffusion pumps benefit from good pumping speed on the lighter elements such as hydrogen and helium. However, there is a risk of oil contamination into the system if the liquid nitrogen cold trap is ever allowed to run dry. In our experience, a suitable turbo-molecular pump can match the helium pumping speed of the oil diffusion pump and provides the best solution. Our system uses a scroll pump to back the turbo-molecular pump and for initial rough-pumping of vacuum volumes. The vacuum system is therefore oil-free, eliminating the possibility of back-streaming oil contamination of the multi-layer thermal insulation surrounding the liquid helium reservoir. The vacuum system is protected in the event of a

mains power supply failure by an automatic gate valve, which isolates the cryostat from the pumping system and prevents venting of the volumes. A VACSCAN mass spectrometer mounted external to the refrigerator is used for leak checking.

A Leybold sealed rotary pump circulates the <sup>3</sup>He/<sup>4</sup>He mixture with an oil mist filter to reduce oil contamination of the mixture and insert. The 1 K pot is pumped with a smaller Leybold rotary pump. Currently, all pumps are located close to the refrigerator on vibration isolation mats. This is not usual practice in this field as pumps are a likely source of acoustic and electromagnetic noise. Most similar systems incorporate pumps in an adjacent room but restrictions on laboratory space made this an impractical solution in our case. If detector resolution becomes limited by noise from the pumps we have measures in hand to reduce this noise: lowering the pumps onto the concrete foundations, constructing electromagnetic shielding and incorporating vibration dampers on the pumping lines to reduce vibration transmitted directly to the refrigerator.

# **3.1.5 Support Structure**

The refrigerator is mounted on a support frame (figure 3.2) designed to reduce mechanical vibration to the refrigerator, originating primarily from the vacuum pumps. To achieve this, the structure incorporates an aluminium table (186 kg) on which the refrigerator (and later the beamline) is mounted. This table is de-coupled from the main support frame by 4 pneumatic mounts. The support frame also provides a useful structure from which to suspend the refrigerator during assembly and disassembly.

#### **3.2** The Superconducting Quantum Interference Device (SQUID)

#### **3.2.1 SQUID Principles**

A dc SQUID is used to readout the TES detector. A dc SQUID consists of two parallel Josephson junctions in a superconducting loop. A Josephson junction [Josephson, B., 1964] is a small separation (on the scale of the coherence length) in the superconductor where the superconductivity is weakened and the critical current reduced. An rf SQUID has just one junction. When a bias current,  $I_b$  is applied to a SQUID, the voltage across the SQUID is zero if the bias current is less than the critical current,  $I_c$ . In the case of a dc SQUID, when the bias current exceeds the critical current, a finite voltage difference across the junctions is produced. This is not the case for an rf SQUID which has a strongly coupled superconducting short circuit across the single Josephson junction. If an external magnetic field is also applied to a dc SQUID, the total critical current (and hence voltage) varies periodically with the magnetic flux quantisation (figure 3.4). The magnetic field is measured by monitoring this

change in voltage. As is clear from figure 3.5, the applied magnetic flux is a measure of the current drawn by the TES, and thus provides a measure of the change in resistance of the TES due to an X-ray photon absorption event.



Figure 3.4: Variation of voltage, V with magnetic flux, with period equal to one flux quantum,  $\Phi_0$ . Peak-to-peak voltage swing is small: typically of order  $\mu V$ .  $d\Phi$  is the flux corresponding to peak voltage.

The voltage response has a period of one flux quantum,  $\Phi_0 = \frac{h}{2e} = 2.07 \times 10^{-15}$  Wb. We used an Oxford Instruments broadband SuperSQUID (a non-commercial development device) which has a linear response for a region of approximately 0.2  $\Phi_0$ , corresponding to a current of 0.6  $\mu$ A. The current may exceed this value when maximum energy (e.g. 6 keV) is incident on the detector and we require a linear output response. Therefore, the SQUID is operated in a negative feedback flux-locked loop (FLL) to linearise the output. A simple example of a FLL circuit is shown in figure 3.5. The SQUID output voltage is amplified, integrated and fed back as a current to a feedback coil which maintains the flux within the SQUID close to zero. This is an example of direct readout electronics where the SQUID and preamplifier are coupled together without an intermediary matching transformer. This is only possible for high values of the gradient of the flux-to-voltage transfer function,  $\frac{dV}{d\Phi}$ . High bandwidths may be achieved (5 MHz), determined by the transmission line characteristics of the feedback circuit and the cut-off frequencies of the preamplifier and integrator [Drung, D., 1995].



**Figure 3.5**: Direct readout FLL electronics, including a low-noise preamplifier, integrator and feedback resistors R1 and R2.

# 3.2.2 Comparison of SQUIDs and Conventional Amplifiers

SQUIDs have many advantages over junction field-effect transistors (JFETs) for low temperature applications. SQUIDs can have high bandwidth ( $\leq$  5 MHz at 3-dB) and produce relatively high output voltages (~ mV) so they can be read out with room temperature op-amps. If room temperature preamplifiers are used there can be problems with microphonics and with the large capacitance of the (long) connecting leads which adds noise and degrades the rise time of the signal. SQUIDs can be mounted close to the detector as they dissipate less power than JFETs ( $\mu$ W rather than mW) and can operate at mK temperatures rather than 77 K for JFETs [Moseley, S., 1984]. This is important in our cryostat because the liquid nitrogen is in a separate vacuum chamber to the detector. Camin, D. et al. [1994] have developed GaAs FETs which can operate below 4 K, but GaAs is known for its 1/f noise [Booth, N., 1996]. Moreover, SQUIDs are easily impedance-matched with low resistance (R ~ 0.1  $\Omega$ ) TES devices.

# **3.2.3 SQUID Parameters**

SQUID performance must be matched to the requirements of the X-ray microcalorimeter. Critical parameters are SQUID noise, dynamic range and speed. In general, the input current range for the SQUID (the current applied to the input coil shown in figure 3.5),  $dI_{in}$  is given by [Frericks, M., 1998]:

$$dI_{in} = \frac{d\Phi(1+A\beta)}{M_{in}}$$
(3-1)

where  $M_{in}$  is the mutual inductance of the input coil. A $\beta$  is the loop gain of the system.  $A = \left(\frac{dV}{d\Phi}\right)G$ 

where G is the complex transfer function of the preamplifier and the integrator, and  $\beta = \frac{M_{fb}}{R_{fb}}$  where  $R_{fb}$  is the total feedback resistance and  $M_{fb}$  is the mutual inductance of the feedback coil.  $M_{fb} = k (L_{fb} L_{sq})^{\frac{1}{2}}$  where  $L_{fb}$  and  $L_{sq}$  are the inductance of the feedback coil and SQUID respectively, with a coupling parameter,  $k \sim 1$ .

The input current noise,  $i_n$  is given by [Gallop, J., 1991]:

$$i_n = \frac{\Phi_n}{M_{in}} \tag{3-2}$$

where  $\Phi_n$  is the flux noise. This is an inherent SQUID parameter and may be approximated by [Frericks, M., 1998]:

$$\Phi_n = \left(\frac{16K_B T L_{sq}^2}{R_{sh}}\right)^{\frac{1}{2}}$$
(3-3)

where  $K_B$  is the Boltzmann constant, T is the operating temperature and  $R_{sh}$  is the shunt resistance.

In the white noise region ( $\geq 1000$  Hz), our Oxford Instruments SuperSQUID has a flux noise spectral density of 0.7  $\mu \Phi_0/\text{Hz}^{1/2}$  and a mutual input inductance,  $M_{in} = 720$  pH [Polushkin, V., 1997]. This corresponds to an input current noise of ~ 2 pA/Hz^{1/2} (measured at 4.2 K). From inspection of equations (3-1) and (3-2), the ratio of input current range to current noise is determined by  $\frac{d\Phi}{\Phi_n}$ .

The minimum dynamic range of the SQUID, f is determined by the requirement to follow the rise time of the detector signal,  $\tau_{rise}$ :

$$f \ge \frac{1}{2\pi\tau_{rise}} \tag{3-4}$$

Our SQUID has a bandwidth of 1 MHz and a typical rise time is of order 1  $\mu$ s for a single pixel device of side length ~ 250  $\mu$ m (or ~ 10  $\mu$ s for an extended bismuth absorber, see Chapter 9). Therefore, the electronics must operate approximately 5 times faster than the thermal response of the detector to facilitate the use of rise time discrimination for background signal rejection. Table 3.1 gives a summary of the performance of this SQUID in comparison with other commercially available SQUIDs. The Hypres SQUID is widely used, in part due to its very large value for  $\frac{dV}{d\Phi}$ . The Oxford Instruments SQUID has high flux at maximum voltage,  $d\Phi$  and thus matches the Hypres SQUID in terms of the basic SQUID quality parameter  $\frac{d\Phi}{\Phi_n}$ , important for low current noise operation.

	Hypres	APF	DROS	Oxford Instruments
$dV/d\Phi$ (mV/ $\Phi_0$ )	31	0.7	1	1.6
$d \Phi(\Phi_0)$	0.16	0.086	0.06	0.2
M <sub>in</sub> (pH)	660	400	5200	720
$\Phi_n \left( \mu \Phi_0 / \text{Hz}^{1/2} \right)$	0.65	3.5	5	0.7
$I_n$ (pA/Hz <sup>1/2</sup> )	2	18	2	2

**Table 3.1**: Summary of the specifications of the most widely used SQUIDs, updated from Frericks, M. et al. [1998]. The Hypres SQUID is manufactured by Hypres Inc. (175 Clearsbrook Rd., Elsmford, NY 10523, USA) and the SuperSQUID is from Oxford Instruments Superconductivity (Newton House, Cambridge Business Park, Cowley Road, Cambridge CB4 4WZ, UK). Also shown for comparison are typical parameters for a SQUID using Additional Positive Feedback (APF) and a Double Relaxation Oscillation SQUID (DROS). APF [Drung, D., 1990] and DROS [Adelerhof, D., 1995] are methods of increasing the flux-to-voltage transfer function,  $\frac{dV}{d\Phi}$  at the working point.

#### 3.3 Packaging Considerations

In parallel with our investigations into the X-ray performance of TESs, we have considered, in the laboratory context, some of the issues regarding the packaging of a detector as a space instrument. These include the physical arrangement of the sensor and SQUID at the cold stage and the method of maintaining good thermal contact between the silicon substrate of the detector and the cold bath. The latter has been our priority at this early experimental stage of detector development. A reliable bond for this silicon-copper interface is a necessary requirement for both single pixel and imaging detectors. Other constraints include high thermal conductivity and low transmission of stress to the substrate when cooled from 300 K to 50 mK. One method is to clamp the silicon to the copper base. However, an adhesive bond allows for greater freedom of movement during cooldown. This compensates for the large differential contraction of silicon and copper (figure 3.6) and transmits less stress to the detector structure. For this reason, an adhesive that remains plastic at low temperature is advantageous. An

additional problem when bonding copper to silicon occurs because silicon has a negative value for the coefficient of thermal expansion in the temperature range 18 - 120 K. This may be approached qualitatively by consideration of the Gruneisen parameter,  $\gamma$  defined as [Rosenberg, H., 1963]:

$$\gamma = \frac{-\partial(\log v_{\max})}{\partial(\log V)}$$
(3-5)

The Gruneisen parameter describes the volume, V dependence of the maximum frequency of atomic oscillation,  $v_{max}$  and may be considered as a measure of the anharmonicity of the vibrations. For a string of atoms in one dimension (x),  $\gamma$  consists of a weighted average of terms,  $\gamma$ , for both the transverse and longitudinal modes. These weightings are a function of temperature and represent the contribution of each mode to the heat capacity. In the case of silicon between 18 - 120 K, the contribution from the transverse modes (negative  $\gamma$ ) exceeds that of the longitudinal modes (positive  $\gamma$ ) leading to a negative Gruneisen parameter and contraction in the x direction upon heating [Ernst, G., 1998]. Conceptually, as the temperature increases the average distance between atoms increases due to increasing vibration in that direction (longitudinal), but is decreased to a greater extent in the orthogonal direction (transverse). How this one-dimensional model of lattice spacing translates to a three-dimensional sample of silicon is yet to be explained [Okazaki, A., 1996], but the results of Okazaki et al. [1996] indicate that the effect is anisotropic and highly dependent on lattice orientation.



**Figure 3.6**: Coefficient of linear thermal expansion,  $\alpha \left(\alpha = \frac{1}{L_{293}} \left(\frac{dL}{dT}\right) 10^{-6} \text{ K}^{-1}$ , where  $L_{293}$  is the length at 293 K) for silicon [Touloukian, Y., 1977] and copper [Touloukian, Y., 1975].

Note the negative value for  $\alpha$ (Si) between 18 K – 120 K, indicating expansion on cooling in this range, in contrast to the rapid contraction of copper.

#### 3.3.1 Adhesive Reliability Testing at 77 K

The simplest method to evaluate the comparative reliability of various adhesives at low temperatures is to cool samples in liquid nitrogen. As can be seen from figure 3.6, most of the thermal contraction of the key materials occurs above 77 K. Thus, although not as rigorous a test as cycling from 293 K to 4 K, thermally cycling to 77 K does enter the regime of negative  $\alpha$  for silicon and was expected to generate representative stresses across the adhesive. Such thermal stress may lead to failure of the bond, or if it is imparted into the silicon substrate, failure of the silicon nitride membrane on which the detector is mounted. The most reliable adhesives are considered to be those with a low value for the glass-transition temperature (the minimum temperature at which the adhesive can undergo plastic deformation). Any differential material contraction resulting from cooling below this temperature induces thermal stress across the adhesive layer.

# **3.3.1.1 Experiment Description**

Four samples of silicon representative of the detector die, 12 mm x 17 mm, were cut from a wafer surplus to the Leicester CCD detector programme and bonded with different adhesives to a length of copper (figure 3.7). A simple aluminium cage, with holes to facilitate free flow of liquid nitrogen, was secured around the copper rod to ensure no mechanical shocks were imparted to the samples. The assembly was lowered into liquid nitrogen. Sample temperature was measured using a Platinum Resistance Thermometer (PRT). After 10 minutes immersion, the copper rod was withdrawn and allowed to warm naturally to above 0 °C. This cycle was repeated 5 times with the first set of samples, and 50 times for the second. The adhesive bonds were checked visually at each cycle.

The first set of 4 samples were bonded with Eccosil 4952 (with primer, cured at room temperature), GE 7031 Varnish (cured at room temperature), NuSil CV1142 (cured at room temperature) and Ablebond 84 1LMI (cured for 1.5 hours at 150 °C). These were subjected to 5 thermal cycles. Adhesive suppliers and their contact addresses are given in table 3.2.

The second set of 4 samples repeated the tests of (a) Eccosil 4952 (with primer, cured at room temperature), (b) NuSil CV1142 (cured at room temperature), and (c) Ablebond 84 1LMI (cured for 2 hours at 125 °C). The second set also included Dow Corning Hansil silicone rubber sealant. The second sample set were subjected to 50 thermal cycles.

The adhesives were chosen for the following reasons. Eccosil 4952, NuSil CV1142 and Dow Corning Hansil are all silicone rubber based, in order of decreasing thermal conductivity. Silicone rubber has a relatively low glass transition temperature ( $\sim -120$  °C) and the cryogenic research group at SRON, Utrecht have had success with mounting their detectors with NuSil CV1142. CV1142 has also been

successfully thermally cycled from -100 to +80 °C for a total of 2000 cycles by L'Garde Inc. of Tustin, California during testing of a lightweight inflatable solar array [Williams, G., 1993]. GE Varnish is routinely used as an adhesive for a range of low temperature (4 K) applications and Ablebond 84 1LMI was used to bond CCDs in the EPIC instrument, flown on XMM.



**Figure 3.7**: 4 Silicon samples, approximately 1 cm<sup>2</sup> in area, mounted with various adhesives onto a copper rod, shown after repeated thermal cycling to 77 K. Note the disrupted silicon of the fourth sample, bonded with Ablebond 84 1LMI.

Adhesive	Supplier	Address
Eccosil 4952 & Ablebond 84 1LMI	Emerson and Cuming	Windsor Court, Kingsmead Business Park, London Rd, High Wycombe, Bucks HP11 1JU
GE 7031	OI Superconductivity	Tubney Woods, Abingdon, Oxon OX13 5QX
NuSil CV1142	NuSil Technology	1050 Cindy Lane, Carpinteria, CA 93013, USA
Silicone rubber sealant	Dow Corning Hansil	19 Wintersells Rd, Byfleet, Surrey, KT14 7LH
EPO-TEK H20E	Epoxy Technology Inc.	14 Fortune Drive, PO Box 567, Billerica, MA 01821, USA

Table 3.2: List of suppliers of adhesives used in this experiment.

# 3.3.1.2 Results

The results are summarised in table 3.3. All of the adhesives remained attached, with the exception of GE Varnish. Most interestingly, during both trials the Ablebond 84 1LMI sample was severely disrupted through the plane of the silicon, although at no point did the bond fail (shown in figure 3.7). This is attributed to efficient transmission of thermal stress induced by differential thermal contraction

of the copper and silicon and is the same effect that motivated us to redesign the detector mounting, as described in section 3.3.2.

Adhesive	Number of thermal cycles before bond failure			
	Trial 1 (5 cycles)	Trial 2 (50 cycles)		
Ablebond 84 1LMI	2	1		
GE Varnish	5	Not tested		
Eccosil 4952	No failure	No failure		
NuSil CV1142	No failure	No failure		
Dow Corning Silicone	Not tested	No failure		

**Table 3.3**: Summary of adhesive thermal cycling trials.

# 3.3.2 Testing of Modified Mounting at 4 K

Our original detector mounting arrangement involved attaching the silicon substrate directly onto a 30 mm square gold plated copper base. A PCB overlayed the silicon (with ~ 1 mm clearance) and short wirebonds were made between the PCB and bond-pads on the chip. A silicone rubber based adhesive (Eccosil 4952) which performed well in the thermal cycling trials to 77 K (section 3.3.1) was applied in two areas between the silicon and copper base. This adhesive also has relatively good thermal conductivity (1.1 Wm<sup>-1</sup>K<sup>-1</sup>). However, when cooled to < 30 K, the accumulated thermal stress across the adhesive was sufficient to fracture the silicon wafer and thereby destroy the silicon nitride membrane. This unexpected result revealed how the initial testing to 77 K was, in this instance, insufficient to fully evaluate the performance of the bond. It also highlighted flaws in this mounting arrangement. Therefore, the mounting scheme was revised and all future tests were conducted to 10 K using a liquid helium cooled cryostat (figure 3.9).

We devised an alternative bonding scheme for near-term laboratory use, shown in figure 3.8. In this design, the silicon wafer is bonded laterally, at four points around the side of the wafer. At each bonding point the copper mounting is machined to flex slightly to compensate for differential thermal contraction. Resultant forces are transmitted through the plane of the silicon and are less likely to flex the silicon. Copper fingers extend under the wafer to ensure adequate thermal contact between the detector and cold bath. With a reduced contact area between the chip and the mounting, thermal conductivity across the bond becomes the key driver in choice of adhesive. Hence, a silver-loaded epoxy (EPO-TEK H20E) was used as the adhesive to maximise thermal conductivity (2800 Wm<sup>-1</sup>K<sup>-1</sup> at 0 °C, over 2000 times greater than Eccosil 4952). EPO-TEK H20E is designed for use at 300 – 400 °C (but is also rated down to –60 °C) and has a high glass transition temperature (50 °C) but this is no

longer a significant disadvantage since flexing of the copper mounting during cooling compensates for the rigidity of the bond.



Figure 3.8: Alternative detector mounting scheme, shown from above and below. The four bonding points are clearly visible on both sides of the silicon, of side length 15 mm. The copper was plated with 2  $\mu$ m of gold for reliable thermal contact.

Three membrane samples were commissioned from Oxford Instruments. To determine if the membrane failed during cooling, each had a gold track deposited across the membrane and electrical continuity was monitored. All three samples were cooled simultaneously in a small liquid helium cryostat (see figure 3.9). Thermometry was provided by a Platinum Resistance Thermometer (PRT) and a calibrated 2200  $\Omega$  ruthenium oxide (RuOx) thermometer. The PRT was calibrated down to 77 K and provides a reasonable estimate down to helium temperatures. The RuOx sensor was calibrated for T < 4 K only, but was sensitive to temperature changes below 10 K. We were able to repeatedly achieve a base temperature of approximately 10 K, which was sufficient to explore the regime of maximum thermal stress (figure 3.6).



**Figure 3.9**: Liquid helium cryostat used for testing to 10 K. The three membrane samples with overlaid PCBs are shown mounted to the base plate of the liquid helium reservoir.

The samples were thermally cycled from 10 K to room temperature three times over a period of two days. All three samples remained electrically continuous for the duration of the test and no visible changes to the samples were evident when inspected under a microscope, indicating that the alternative mounting design was working correctly. All subsequent detectors were mounted using this arrangement and no failures attributable to thermal stress damage have occurred. With this phase of the programme complete, we were able to concentrate on optimising the detector design, the details of which are described in Chapter 5.

# **Chapter 4**

# Low temperature MCP testing

# 4.1 Microchannel Plate Electron Multipliers

The low temperature testing techniques developed for the TES programme have also been applied to other detector programmes at Leicester. Microchannel plates (MCPs) are high gain electron multipliers consisting of ~  $10^7$  close-packed channels of common diameter, D ( $D \sim 10 \mu m$ ), each operating as an independent photomultiplier [Fraser, G., 1989]. The MCP may therefore be used as an imaging detector of charged particles or high energy photons, most often at room temperature. The performance of MCPs at cryogenic temperatures, however, is also of great interest. An MCP detector is operated at 70 K in the Los Alamos antiproton gravity experiment [Schecker, J., 1992] and an MCP at  $T \leq 20$  K has been proposed as the readout element for a novel liquid helium-based quantum computer [Platzman, P., 1999].

MCPs are fabricated from lead glasses, with principal constituents PbO and SiO<sub>2</sub>. The high lead content gives the hard X-ray response, soft X-ray interaction is with a thin (~ 10 nm) silica-like surface layer which is lead-depleted but potassium (and carbon) enriched. The secondary electron yield of this activated layer determines the MCP gain [Trap, H., 1971] and compositional changes in this layer are largely responsible for changes in plate resistance, at a given temperature.

The resistance, R of an MCP is an important parameter for low temperature applications where levels of heat dissipation may be critical. In addition, the count rate capacity depends on the effective RC channel recharge time constant [Fraser, G., 1991]. The electrical resistance of any MCP increases with decreasing temperature in the manner of a semiconductor. It follows, therefore, that the room temperature resistance may lead to an estimate of the performance at low temperatures, once the R(T)curve is understood. The experiment described in this Chapter extends the work of Roth, P. and Fraser, G. [2000b] in which results from an initial study of low temperature MCP resistance were presented.

The low resistance microchannel plate used for this experiment was supplied by Nova Scientific Inc., Sturbridge Business Park, 660 Main Street, PO Box 928, Sturbridge, MA 01566, USA. Previous experiments were conducted on a plate fragment with room temperature resistance R(293 K) = 4.6MQ [Roth, P., 2000b]. This increased by a factor of 10<sup>6</sup> when cooled to 20 K, potentially too high for use in a quantum computer application. The  $\frac{R(T)}{R(293K)}$  curve was modelled but the validity of the result to plates of different room temperature resistance remained an open question. We have tested an MCP fragment with room temperature resistance  $R(293 \text{ K}) = 200 \text{ M}\Omega$  - a factor of 40-50 times higher than that of the original plate - to further investigate the physics involved in changes in MCP resistance with temperature. It may be possible to manufacture plates with room temperature resistances as low as 10 k $\Omega$  which should be well suited to cryogenic applications. We intend to test such plates when they become available.

## 4.2 Experiment Description

Electrical connections to the MCP were made using contacts on the top and bottom faces of the plate, bonded with EPO-TEK H20E silver loaded epoxy, as used for mounting TES detectors (see Chapter 3). The room temperature resistance was taken to be the mean value determined by the application of dc voltages in the range 1-25 V (to ensure self heating was not affecting the results). Current was measured using a Keithley Instruments solid state 602 electrometer, sensitive to 10 pA. Cooling of the plates was achieved by fixing the plate to the end of a stainless steel tube and inserting the assembly into a small (~31) liquid helium dewar. When the top of the partially filled dewar was sealed, temperature stratifications were established above the helium surface, so by moving the plate vertically it was possible to cover the required temperature range. Resistance measurements during cooling were taken using an applied dc voltage of 20 V. The temperature was measured with a Pt resistance thermometer located close to the MCP.

#### 4.2.1 Results and Discussion

Figure 4.1 is a plot of the resistance of the MCP, scaled to its room temperature value, as a function of temperature.



**Figure 4.1**: MCP resistance normalised by the room temperature value (200 M $\Omega$ ). Shown in red for comparison are data taken by Roth [2000b] of an MCP with a room temperature resistance of 4.6 M $\Omega$ . Each data set is modelled by a curve of the form suggested by Roth [2000b].

The cooling curves for two MCP fragments are shown: the new data with room temperature resistance of 200 M $\Omega$  and the other, previously measured at 4.6 M $\Omega$  [Roth, P., 2000b]. The form of the best-fit curve to the latter is given by Roth (henceforth called 'Model 1') and consists of two components:

$$\frac{R_{\rm I}(T)}{R(293)} = e^{\left(\frac{270}{T}\right)} \qquad \text{for } 4 < T < 90 \text{ K}$$

$$\frac{R_2(T)}{R(293)} = 0.3e^{\left(\frac{380}{T}\right)} \qquad \text{for } 90 < T < 293 \text{ K}$$

$$(4-1)$$

To determine an equivalent curve for our MCP data, the logarithm of the resistance ratio was plotted as a function of inverse temperature (figure 4.2).



Figure 4.2: Arrhenius plot of the resistance ratio against inverse temperature. A linear fit is assumed.

Discontinuities in the data coincide with changes in scale on the Keithley ammeter; reading errors due to variation of the analogue response (estimated to be ~ 5 %) scale with decreasing current. Figure 4.2 is best described by a single linear function, the gradient and intercept of which yield the form of the best-fit curve shown in figure 4.1 ('Model 2'):

$$\frac{R(T)}{R(293)} = 0.32e^{\left(\frac{306}{T}\right)} \quad \text{for } 0 < T < 293 \text{ K}$$
(4-3)

This differs from Model 1, which implicitly assumes that two conduction mechanisms are involved, with activation energies of 23 meV and 33 meV. From equation (4-3), the single activation energy is equal to  $\frac{306}{K_B} = 26$  meV, where  $K_B$  denotes Boltzmann's constant. It is possible that the single conduction mechanism of Model 2 is simply an approximation to the double conduction mechanisms of Model 1, particularly given the errors in the resistance data and the fact that the single activation energy lies approximately mid-way between values of Model 1.

An important point to note from figure 4.1 is the convergence in modelled curves at low temperatures (T < 50 K). This indicates that the  $\frac{R(T)}{R(293)}$  curve is generic and suggests that an MCP with a room temperature resistance four orders of magnitude lower than studied so far would have a similarly reduced resistance at low temperatures (i.e.  $R(293) = 10 \text{ k}\Omega$  rather than ~ 100 M $\Omega$ ). Figure 4.3 compares the two forms of modelled fit as a function of temperature.



**Figure 4.3**: Plot of the ratio of the normalised resistances predicted by Model 1 and Model 2, as a function of temperature. The impact of the change in profile of Model 1 at 90 K is clear.

The room temperature resistances of the MCPs on which these models are based differed by a factor of approximately 50, yet agreement between the two models is good: to ~ 50 % or better over the temperature range 20-300 K. Therefore, we are confident that a low resistance MCP (10 k $\Omega$ ) would behave in a similar manner at temperatures below 20 K and hence may be suitable for use in a quantum computer.

Then:  $R(20K) \approx 10^6 \times 10 k\Omega = 10^{10} \Omega$ 

The physics behind the change of resistance with temperature remains unclear. Ionic conductivity is unlikely to dominate at low temperatures. An alternative mechanism discussed in Roth [2000b], is the variable range hopping model used by Sandoe et al. [1980]. Their data for a higher resistance (2100 M $\Omega$ ) MCP was well described by the conductivity-temperature relation [Mott, N., 1969]:

$$\sigma(T) = \sigma_0 e^{\left(-\left(\frac{T_0}{T}\right)^n\right)}$$
(4-4)

where  $\sigma_0 = \left(\frac{C}{T}\right)^{\frac{2}{3}}$ , C is a constant, and  $n = \frac{1}{3}$  for 2-D conduction and  $n = \frac{1}{4}$  for 3-D conduction.

Following Roth [2000b], plots were made of  $\ln\left(RT^{-\frac{2}{3}}\right)$  versus  $T^{-\frac{1}{3}}$  and  $T^{-\frac{1}{4}}$ . A straight line in either

case would indicate the presence of a particular hopping mechanism. Figure 4.4 shows a plot for the

present data for the 3-D case. Significant non-linearity can be seen in the data. If a linear fit is assumed, then regression yields an  $r^2$  coefficient of only 0.968. The equivalent analysis for the 2-D case is slightly more linear and yields an  $r^2$  coefficient of 0.975.



Figure 4.4: Plot to investigate consistency of the R(T) data with the 3-D variable range hopping mechanism. Non-linearity indicates a poor fit to this model.

A further conduction model is given by Gzowski et al. [1982], who modelled the MCP surface as an array of lead crystallites of common diameter equal to their average spacing. The conductivity is then given by:

$$\ln \sigma(T) = DT^{-\frac{1}{2}} \tag{4-5}$$

When we plot  $\ln(R)$  as a function of  $T^{-\frac{1}{2}}$ , the result is of a similar form to that of figure 4.4 with an  $r^2$  coefficient of 0.972, very close to the value for 2-D hopping.

# 4.3 Conclusion

These inconclusive results are very similar to those obtained by Roth [2000b]. We can therefore corroborate the conclusions drawn therein, particularly with regard to the fact that the 2D hopping conduction mechanism in an MCP is probably similar to that commonly seen in amorphous semiconductors. Our high resistance data set does not permit determination of the power-law exponent of temperature. However, and more importantly, our results endorse the view that the R(T) curve follows a universal profile irrespective of the room temperature resistance, with promising implications for the development of a quantum computer.

# **Chapter 5**

# **Detector design and fabrication**

#### **5.1 TES Design Constraints**

The physical dimensions of a TES are primarily determined by thermal conduction and electrical resistance which are material dependent parameters. For example, the maximum length of a TES may be set by the requirement for both ends of the TES to be at a similar temperature. Moreover, to avoid superconducting/normal phase separation, the temperature difference along the TES must never be greater than the superconducting transition width. Therefore, the maximum bias power is determined by the thermal conductivity of the TES material and varies with TES dimensions.

If it is stipulated that the bias power is dissipated uniformly along the length of the TES, then, per unit length, l the applied bias power must satisfy:

$$\frac{P_{bias}}{l} \ll G_{wf} \Delta T \tag{5-1}$$

Where  $G_{wf}$  is the thermal conductivity given by the Wiedemann-Franz relationship  $(G_{wf} = \frac{L_0 T}{\rho}]$ , where the Lorentz number,  $L_0 \sim 2.45 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$  [Walton, A., 1983],  $\rho$  is the resistivity) and  $\Delta T$  is

$$\Delta T \approx \frac{T_c}{\alpha} \tag{5-2}$$

For a single TES under dc bias [Labov, S., 1997]:

the width of the superconducting transition:

$$P_{bias} = \Sigma A l_{\max} \left( T_e^n - T_b^n \right)$$
(5-3)

Where  $\Sigma$  is the electron-phonon coupling parameter. This is material dependent but typically lies in the range 1 x 10<sup>8</sup> – 3 x 10<sup>9</sup> Wm<sup>-3</sup>K<sup>-5</sup> [Goldie, D., 1999].  $l_{max}$  is the maximum length of the TES, A is the cross section area of the TES (as shown in figure 5.1) and n = 5 for electron-phonon coupling. The temperature subscripts e and b refer to the electron and bath temperatures respectively.



Figure 5.1: Diagram of TES to illustrate the thickness, d, length,  $l_{max}$ , width, w and cross section area A referred to in the text.

Since the conductance between electrons and phonons in the TES,  $G_{e\phi}$  is given by [Wellstood, F., 1994]:

$$G_{e\phi} = \sum nAl_{\max} T_e^{n-1}$$
(5-4)

It follows that:

$$P_{bias} = \frac{G_{e\phi}T_e}{n} \left(1 - \left(\frac{T_b}{T_e}\right)^n\right)$$
(5-5)

In the limit of low bath temperatures ( $T_b \ll T_e$ ) and evaluated at the transition temperature, this simplifies to:

$$P_{bias} \approx \frac{G_{e\phi}T_c}{n} \tag{5-6}$$

Substituting (5-6) and (5-2) into (5-1), it follows that the maximum normal resistance is:

$$R_{n_{\max}} = \frac{L_0 T n}{G_{\alpha \alpha} \alpha}$$
(5-7)

From the definition of resistivity, an alternative expression for  $R_n(max)$  is:

$$R_{n_{\max}} = \frac{\rho l_{\max}}{A} \tag{5-8}$$

Equating equations (5-5) and (5-6) and substituting from (5-4) for  $G_{e\phi}$  yields the limit on  $l_{max}$ :

$$l_{\max} = \sqrt{\frac{L_0}{\alpha \rho \Sigma T^{n-2}}}$$
(5-9)

Assuming  $\Sigma = 0.5 \times 10^9 \text{ Wm}^{-3}\text{K}^{-5}$  (an estimate for the lower limit for  $\Sigma$ , and hence the maximum TES dimensions [Goldie, D., 2000]), n = 5 and substituting measured values for  $\rho$ ,  $\alpha$  and  $T_c$  from two different deposition runs of iridium test resistors (Chapter 7), yields values for  $l_{max}$  ranging from approximately 40 µm to 1000 µm. The large spread in values in this single parameter is an example of the critical nature of optimum TES design on material parameters.

For a TES of fixed length, A, the product of the width, w and thickness, d, should be maximised in order to minimise the electrical resistance. In practice, for a single pixel detector, the width of the TES is limited by the dimensions of the absorber. Greater flexibility in the width is available in large area absorber designs. In practice, the minimum resistance is determined by how small the bias resistor can be made; typically ~ 1 m $\Omega$ . Hence, for stable operation under voltage bias at a TES resistance of 0.1R<sub>n</sub>, a normal resistance of 100 m $\Omega$  is required. This places an upper limit on the thickness of the TES:

$$d_{\max} = \frac{\rho l_{\max}}{0.1w} \tag{5-10}$$

Hence, the maximum volume of a TES is:

$$vol_{\max} = \frac{L_0}{0.1\alpha\Sigma T^{n-2}}$$
 (5-11)

This may be substituted back into (5-4) to show how the maximum conductance between electrons and phonons varies linearly with temperature:

$$G_{e\phi_{\max}} = \frac{nL_0T}{0.1\alpha}$$
(5-12)

Maximum count rate is primarily determined by pulse length, hence a rapid decay time,  $\tau_{eff}$  is desirable. When operating with electrothermal feedback,  $\tau_{eff} \approx \frac{Cn}{G\alpha}$  (see equation (2-3)), where C is the total specific heat capacity of the detector (TES plus absorber and wiring) and G is the thermal conductivity to the heat bath. The heat capacity is largely determined by the dimensions of the absorber, which in turn depend on the required pixel area and quantum efficiency. However, the maximum value for G is constrained by the maximum volume of the TES (equation (5-11)) and the electron-phonon conductance (equation (5-12)).

The derivations above show how the choice of TES material influences the design of the detector and places limits on the device geometry. The results are valid for single material layers and bilayers. The effect of TES material upon the reliability of the device is discussed in chapter 6.

### **5.2 TES Fabrication**

# **5.2.1 Film Deposition Techniques**

All of our TES devices were deposited by physical vapour deposition – either evaporation or sputtering. In evaporation, atoms are transferred from a source to a substrate by thermal means [Maissel, L., 1970]. In sputtering, atoms are forced from a solid source target by the impact of gaseous ions. The sputtering process is best suited to the growth of alloy films with stringent stoichiometry limits, whereas evaporation is a simpler technique with consequently easier control over the deposition parameters [Berry, R., 1968]. The Ti/Au bilayers fabricated by SRON were deposited by high vacuum electron beam evaporation, but for the iridium detectors fabricated by Oxford Instruments, all wiring and absorbers were deposited by sputtering.

#### **5.2.2 Deposition Conditions**

The performance of a thin film depends critically upon the conditions under which it was deposited, for both sputtering and evaporation techniques. The quality of the vacuum is a key parameter, in particular the background level of oxygen. Highly reactive metals, such as titanium, are often exposed in the vacuum chamber prior to deposition to deplete excess oxygen. However, it is not possible to remove all the oxygen, and the expected rate of oxidation of the sample must be taken into account when setting the deposition rate. The temperature of the substrate defines the size and form of the grains. Heating the substrate increases the surface mobility of depositing atoms, resulting in more uniform coverage [Ohring, M., 1992]. For  $T_{melt}/T_{sub} < 3$ , (where the subscripts refer to the melting point of the metal and substrate temperature) grain size increases with temperature and the grains deposit in closely packed columnar structures [Wagendristel, A., 1994]. For  $T_{melt}/T_{sub} > 6$ , the grain size is independent of temperature and the grains deposit in a porous structure consisting of tapered crystallites separated by voids [Hentzell, H., 1984]. The transition structure consists of densely packed fibrous grains. The temperature also defines the stress in the grown film: for thin films of refractory metals such as molybdenum, it has been shown that a substrate temperature of 800 °C  $(T_{melt}/T_{sub} \sim 3)$  results in minimum film stress [Thornton, J., 1977]. We have experimented with a range of deposition temperatures for our iridium films (see Chapter 7) to investigate the effects on superconducting-normal transition properties.

# 5.2.3 TES Processing

#### 5.2.3.1 Patterning

The patterning of the detectors was achieved by either negative photolithographic lift-off techniques or ion beam milling (IBM). In IBM, argon ions formed by an electrical discharge are accelerated towards a target by a pair of optically aligned charged grids. The target is mounted on a rotating, temperature controlled workplate with a neutralising filament to protect against the accumulation of positive charge. The incident ions remove any metal not protected by a resist material. A similar result is obtained by using lift-off processes. In this case, a negative mask is brought into intimate contact with the substrate and a layer of photoresist is applied to the exposed surface. The mask is removed and the required metal deposited over the entire surface by evaporation or sputtering. The photoresist is stripped by exposure to acetone, thereby leaving only the desired metal pattern which is a negative of the resist pattern (figure 5.2).



Figure 5.2: Three stages for patterning by negative lift-off photoresist method.

#### 5.2.3.2 Processing Steps

Details of the steps for processing a TES detector are generally considered confidential and vary between laboratories but it is possible to give a brief outline of a typical procedure.

The silicon substrate is cleaned in acetone, rinsed in isopropyl alcohol (IPA) then blown with nitrogen or spun dried prior to a soft bake to remove any moisture from the surface. A spin-coat photoresist layer may then be applied. This is exposed to UV light for approximately 20 seconds before immersion in developer solution. The desired metal may then be sputtered onto the chip, often through a simple shadowmask, and then lift-off of the resist layer is achieved by immersion in acetone for about 30 minutes. Alternatively, an ion beam milling process may be used to remove metal unshielded by resist from the chip surface. The photoresist application and lift-off process is repeated for each layer of the device: the TES material (Ir, Ti, Au), wiring (Al, Nb), shunt resistors (Ag) and absorber (Cu, Au). The masks may incorporate test layers and structures on the chip (but not on the membrane) so that the quality of the detector elements may be monitored. Once all components of the detector are deposited, the silicon is anisotropically back-etched by immersion in a potassium hydroxide (KOH) solution for a controlled time to produce the membrane (thickness > 100 nm). When the required depth of etch has been reached the chip is rinsed and mounted onto a copper holding plate with a suitable adhesive.

#### 5.3 On-chip Test Structures

The detectors made under contract to Leicester by Oxford Instruments include several on-chip test structures for materials analysis.

#### **5.3.1 Material Interface Stability**

An important consideration for the reliable operation of a detector is the compatibility of materials that constitute each component of the device, and test structures have been deposited to investigate this. The phase diagrams in the literature yield much information about each interface. The conclusions may be summarised as in table 5.1.

Interface	Application	Comments
Ir – Au	TES – absorber	Immiscible
Ir – Nb	TES – bias rails	Intermetallics
Ir – Ag	TES – bias rails	Intermetallics – no available data on ageing
Au – Al	absorber – bias rails	Intermetallics
Au – Nb	absorber – bias rails	Intermetallics
Ag – Nb	Shunt resistor – bias rails	Immiscible

**Table 5.1**: Nature of metallic interface systems [Moffatt, W., 1986]. Systems with solid solubility of less than 1 % at room temperature are described as immiscible. This takes no account of possible effects due to elevated temperatures during deposition or processing.

The presence of intermetallics may not necessarily compromise the performance of the detector, depending on the rate of intermetallic formation and how the new compounds effect the contact resistance across the material interface. The Ag/Al/Ag trilayer provides an example of a detector configuration which is seriously compromised by intermetallic effects, and is described in Chapter 6. Data on ageing of these systems and the effects of thermal cycling are limited. The current Oxford

Instruments design uses a Au absorber in contact with an Ir TES with Nb tracking, but if degradation (increased series resistance) is observed then it is possible to incorporate Al bias rails in future. The development of interface intermetallics may be monitored via changes in contact resistance. Each material pair (Nb – Ag, Nb – Ir and Nb – Au) has been deposited on detector chips as a series connection of 108 20  $\mu$ m x 20  $\mu$ m squares (figure 5.3).

No significant changes have been observed after storage for periods of up to 12 months at room temperature. Experiments are planned to monitor resistance at elevated temperatures to generate Arhhenius plots and therefore determine an estimate for the ageing rate of these contacts.



**Figure 5.3**: Optical microscope image of the pattern of 108 overlaid squares (of side 20  $\mu$ m) of alternating Ir – Nb, forming a large series resistance sensitive to changes in contact resistance between the materials. Similar patterns have been deposited for Nb – Au and Nb – Ag.

#### 5.3.2 Resistivity Measurements

Separate structures (figure 5.4) have been fabricated for measurement of resistivity of the TES, shunt resistors (Ag) and absorber (Au). The dimensions of the pattern for these test resistors is set to 1619 squares. Ohms per square is a convenient way to refer to sheet resistance. For a rectangular film of length *l*, width *w*, depth *d*, and resistivity  $\rho$ , the resistance is given by  $R = \frac{\rho l}{wd}$ . In the case of a

square film l = w and  $R = R_s = \frac{\rho}{d}$  Ohms per square. The test resistors are 500 nm thick and at this thickness the resistivities of Au and Ag are similar ( $R_{sq} \sim 10 \text{ m}\Omega$ ), resulting in a low temperature resistance of 16  $\Omega$ .



Figure 5.4: On-chip test Ir resistor of thickness 500 nm for resistivity measurements with superconducting Nb tracking.

# **5.3.3 Ir Transition Measurements**

There are also two test iridium structures on-chip to allow for the measurement of transition curves of small iridium devices and to determine the electron – phonon coupling parameter for iridium (equation (5-3)). The test structures are 50  $\mu$ m long and of widths 5  $\mu$ m and 18.75  $\mu$ m, leading to a low temperature, normal state resistance of 1-2  $\Omega$ . The length is limited by the need to avoid phase separation (equation (5-7)). These are visible to either side of the membrane in figure 5.5.

#### **5.3.4 Alignment Features**

The relative alignment of each layer of the mask is achieved either by dedicated features in the masks or by comparison of orientation with existing detector features. Tolerance levels are variable across the detector: TES to membrane alignment of ~ 50  $\mu$ m is sufficient whereas more critical alignment is required for the bias rails to TES/shunt resistors and between the absorber and TES (where the TES extends under the absorber to within 5  $\mu$ m of the edges, see figure 5.6). Alignment features are clearly visible surrounding the membrane in figure 5.5.



Figure 5.5: Sample mask design showing the central membrane with TES and absorber, surrounding test structures and bond pads, superposed with a 1 mm grid to indicate scale. Iridium is shown as white, silver as light blue, niobium as darker blue, gold as yellow and the membrane perimeter as magenta. The two shunt resistors (10 m $\Omega$  and 20 m $\Omega$ ) are shown in the lower right-hand corner. Alignment features encircle the membrane.



Figure 5.6: Optical micrograph of an iridium TES detector on a  $Si_3N_4$  membrane. The edges and overlap of each component are visible, indicating the alignment precision required between masks.

# **5.4 Conclusion**

In summary, the dimensions of the TES (and hence the maximum count rate of the device) are dictated by material properties such as thermal and electrical conduction and the electron – phonon coupling coefficient,  $\Sigma$ .  $\Sigma$  may vary between materials by an order of magnitude, and published data at low temperature is very limited. To optimise the detector design, it is therefore important to include provision for measurement of  $\Sigma$ , determined by the response of iridium test structures of different dimensions to d.c. bias (equation (5-3)). The detector TES could be used for such testing, but at a risk to the device. The fabrication techniques employed for the manufacture of a TES are relatively simple, but cleanliness throughout the process is critical since the operation of the detector depends on the purity of material layers and clean surface interfaces for reliable electrical and thermal contacts. As part of an on-going programme of detector development, we incorporate test structures on each chip to monitor different aspects of material interface reliability.

# **Chapter 6**

# **TES thermometer reliability**

### **6.1 Proximity Layer Reliability**

One concern with the use of proximity layer superconductors (Chapter 2) in a space instrument is the stability of the layers over time. At non-zero temperatures, there is always a degree of interdiffusion between the layers that may lead to alloy formation and significant changes in the transition temperature and profile. This is exacerbated by the elevated temperatures involved during processing of the devices (Chapter 5). The stability of a TES may also be affected by chemical reactions (particularly oxidation) and corrosion occurring during processing of the films, or as a result of exposure to atmospheric oxygen and humidity during storage of the detectors. Resolution of these problems is essential for the development of a space instrument, and has formed a large part of our recent work [Holland, A., 1999].

#### 6.2 Diffusion in Bulk Solids

In any material system that is not in thermodynamic equilibrium, forces arise to reduce free-energy variations (this is particularly important for thin films where compositional inhomogeneities abound at a microscopic level [Gupta, D., 1988]). These forces establish atomic concentration gradients, leading to mass transport by diffusion. In this context, diffusion is defined as the migration of an atomic or molecular species within a given matrix under the influence of a concentration gradient [Ohring, M.,

1992]. The atomic flux J arising only from a concentration gradient  $\frac{dC}{dx}$ , (i.e. no external driving forces such as chemical, thermal or electrical gradients) is given by Fick's law [Shewmon, P., 1989]. In one dimension:

$$J = -D\frac{dC}{dx} \tag{6-1}$$

where D (cm<sup>2</sup>s<sup>-1</sup>) is the diffusion coefficient. The magnitude of D depends on the diffusing species and the medium in which transport occurs and increases with temperature according to a Maxwell-Boltzmann relation:

$$D = D_0 e^{\left(\frac{-Q}{K_B T}\right)} \tag{6-2}$$

Where Q is the excitation energy,  $K_B$  is Boltzmann's constant and  $D_0$  is a constant given by Gupta [1988]:

$$D_{0} = \frac{1}{6} a^{2} z f v_{0} e^{\left(\frac{(s_{f} + s_{m})}{\kappa_{B}}\right)}$$
(6-3)

Where a is the nearest neighbour atomic distance, z describes the probability of finding a suitable defect (lattice vacancy) for the atom to occupy,  $f (\leq 1)$  is a correction term for the non-random nature of the atom jumps,  $v_0$  is the atomic vibration frequency, and  $S_f$  and  $S_m$  are the entropies for the formation and motion of a defect respectively.  $H_f$  and  $H_m$  are the corresponding enthalpy terms and define the excitation energy, Q:

$$Q = H_f + H_m \tag{6-4}$$

A summary of the material parameters most relevant to our investigations is given in table 6.1.

Element	$D_0 (cm^2 s^{-1})$	H <sub>f</sub> (eV)	H <sub>m</sub> (eV)	Q (eV)
Al	0.047	0.67	0.62	1.28
Ag	0.04	1.13	0.66	1.76
Au	0.04	0.95	0.83	1.76
Cu	0.16	1.28	0.71	2.07
W	1.88	3.6	1.8	6.08
Ti	0.00007	•	-	1.75

**Table 6.1**: Lattice self-diffusion (diffusion of atoms within their own lattice) values for selected pure elements [Gupta, D., 1988]. The values for  $H_f$  and  $H_m$  are from Balluffi [1978], whereas the values for Q are from Peterson [1978], hence the slight discrepancy between the sum of  $H_f$  and  $H_m$ , and Q.
#### **6.3 Diffusion in Thin Films**

Polycrystalline thin films differ from the corresponding bulk materials in the proportion of atomdefect combinations associated with grain boundaries, dislocations and surface effects relative to lattice sites. Diffusion processes may be conveniently divided into diffusion through the lattice, including diffusion via lattice dislocations, and diffusion between grain boundaries (see figure 6.1). Fine grain size in thin films means that the activation energies for grain boundary and dislocation diffusion are smaller than those for bulk material, leading to higher total diffusion rates.



**Figure 6.1:** Schematic of the diffusion profile for a Ti-Au system. The curved lines are isoconcentration contours for the diffusion source, Ti in this case. This corresponds to type-B diffusion kinetics in the notation of Harrison [1961], and is intermediate between the extensive lattice diffusion of type-A kinetics and the grain boundary diffusion dominated type-C kinetics.

For a quantitative comparison, consider a polycrystalline film with grains of side length l, grain boundary width  $\delta$ , and dislocations of cross section  $A_d$  occurring within the grains with a density  $\rho_d$ cm<sup>-2</sup>. The number of atoms flowing per unit time for a given concentration gradient  $\frac{dC}{dx}$ , is [Ohring, M., 1992]:

$$n_{l} = D_{l}l^{2} \left(\frac{dC}{dx}\right)_{l} \qquad \text{for lattice diffusion} \qquad (6-5a)$$
$$n_{b} = \delta D_{b}l \left(\frac{dC}{dx}\right)_{b} \qquad \text{for grain boundary diffusion} \qquad (6-5b)$$

$$n_d = A_d D_d l^2 \rho_d \left(\frac{dC}{dx}\right)_d$$
 for dislocation diffusion

where  $D_l$ ,  $D_b$  and  $D_d$  represent the relevant diffusion coefficients.

For face centred cubic (fcc) metals (which include Al, Au, Ag and Cu), the temperature dependences of these parameters are given by Balluffi [1975]:

Lattice diffusion:

$$D_l = 0.5e^{\left(\frac{-17T_M}{T}\right)} \text{ cm}^2\text{s}^{-1}$$

Grain Boundary diffusion:

$$\delta D_{b} = 1.5 \times 10^{-8} e^{\left(\frac{-3.5T_{M}}{T}\right)} \text{ cm}^{3} \text{s}^{-1}$$

$$A_{d} D_{d} = 5.3 \times 10^{-15} e^{\left(\frac{-12.5T_{M}}{T}\right)} \text{ cm}^{4} \text{s}^{-1} \qquad (6-6)$$

(\_80T )

(6-5c)

Dislocation diffusion:

where  $T_m$  is the melting point of the metal ( $T_m = 933$  K for Al and 1234 K for Ag).

Therefore, (assuming typical values for thin films of  $l = 10^{-4}$  cm and  $\frac{T}{T_M} = \frac{1}{3}$ ), the ratio of grain boundary and lattice fluxes is:

$$\frac{n_b}{n_l} = \frac{\delta D_b}{lD_l} = 3 \times 10^{-8} \left(\frac{1}{l}\right) e^{\left(\frac{8.1T_M}{T}\right)} = 1.1 \times 10^7$$
(6-7)

Similarly for dislocation diffusion (assuming  $\rho_d = 1 \times 10^5$  cm<sup>-2</sup> [Ohring, M., 1992]):

$$\frac{n_d}{n_l} = \frac{A_d \rho_d D_d}{D_l} = 1.06 \times 10^{-14} \rho_d e^{\left(\frac{4.5T_M}{T}\right)} = 7.7 \times 10^{-4}$$
(6-8)

Therefore, grain boundary diffusion usually dominates for films with grain sizes  $< 1\mu$ m. Lattice diffusion dominates only in dislocation-free epitaxial films [Ohring, M., 1992]. The flux of both dislocation and grain boundary diffusion relative to lattice diffusion increases with reducing temperature.

From inspection of equations (6-7) and (6-8), the nature of diffusion in our films (when stored at room temperature) is taken to be rapid flow down grain boundaries until such time that all grain boundaries are filled. Then, slower lattice diffusion becomes important. Dislocation diffusion may be neglected.

# **6.4 TES Material Options**

#### **6.4.1 Bilayer Configurations**

Bilayer, or multilayer, devices include normal – superconducting and superconducting – superconducting systems. All of our studies have focussed upon normal – superconducting configurations exploiting the proximity effect at low temperature (Chapter 2).

# 6.4.1.1 Al/Ag

The Al/Ag system is attractive because both materials are easy to fabricate as thin films, and thin film aluminium typically has reproducible superconducting properties. Many groups around the world used this system in early TES detectors, particularly after the first excellent X-ray results using Al/Ag bilayers were published [Irwin, K., 1996]. However, corrosion of the aluminium layer following manufacture, first reported by Finkbeiner et al. [1998], has been a widespread problem. We saw this in samples prepared by colleagues at SRON, Utrecht and Oxford Instruments Superconductivity, Cambridge. The corrosion can take two forms: electrochemical reactions where water from atmospheric humidity acts as an electrolyte, or a chemical reaction enhanced by sulphur which is present in most photoresists. Many photoresists and resist developer solutions also attack aluminium directly. Our initial solution was to fabricate Ag/Al/Ag trilayers which encapsulate the aluminium and protect it from the corrosive effects of processing chemicals and environmental corrosion. These trilayers, which are discussed in detail in section 6.5.4, were still subject to long-term degradation due to interdiffusion of the silver and aluminium. This is the most serious problem with the Al/Ag system, and remains unresolved. We are no longer pursuing the Ag/Al/Ag multilayer approach.

### 6.4.1.2 Mo/Au

The Mo/Au bilayer has been studied by Finkbeiner et al. [1998]. This configuration appears stable and both metals are highly resistant to corrosion. Mo/Au is less diffusive than Al/Ag, with negligible diffusion expected for temperatures less than 300 °C [Smithells, C., 1962]. At temperatures of 200 – 800 °C, the diffusion of molybdenum in gold is reported to be 0.5 - 1% (depending on temperature) [Stahle, C., 2000a] with no diffusion of gold into the molybdenum, but some gold (2 - 3%) may be deposited in the grain boundaries of the molybdenum during film deposition [Hoevers, H., 1999c]. Finkbeiner et al. [1998] report that the superconducting properties of the molybdenum are highly dependent on the substrate and temperature of deposition, although reproducible transitions of width 0.5 mK at about 130 mK are possible. Excellent X-ray results (2.8 eV FWHM at 1.5 keV) have been obtained using TES detectors fabricated from Mo/Au bilayers [Stahle, C., 2000b], and this combination is one of the most promising for future developments.

# 6.4.1.3 Ti/Au

Our colleagues at SRON fabricated all of the Ti/Au bilayers that we have tested. During the development phase of these thermometers, they produced a run of 12 bilayers with layer thickness of the order of 25 nm. These had transition temperatures ranging from 170 - 425 mK and were tolerant of thermal diffusion processes: for example, the transition temperature of one sample changed from 403 mK to 405 mK after storage for 6 months at room temperature. Subsequent testing has enabled SRON to control the transition temperature to accuracies of order 10 mK. Luukanen et al. [1999] at the University of Jyväskylä measured the activation energy for Ti/Au intermetallic diffusion to be 1.18 eV, which they interpret to be a combination of energies for grain boundary and lattice diffusion processes. As is usual for thin films, initial ageing is fast due to grain boundary diffusion and then once the grain boundaries are saturated the slower process of lattice diffusion dominates. Luukanen et al. [1999] calculate a 10 % change in resistance over 15000 years for films at 0 °C. This is only an estimate based on extrapolation of the data, but is indicative of the slow diffusion in this system.

#### **6.4.2 Monolayer Configurations**

In contrast to bilayer configurations, any single material TES has the major advantage of not being subject to interdiffusion related degradation. Reliability concerns reduce to issues of oxidation and adherence/reactions with the substrate. Tungsten and iridium are examples of elements with bulk transition temperatures in the correct range for this application (15 mK and 112 mK, respectively). However, the properties of superconductors are often changed from the bulk values when deposited as a thin film. For example, W and Re typically show an increase in  $T_c$  (from 15 mK to ~ 3 K and from 1.7 K to 7 K, respectively). For these metals there is evidence of the presence of a phase (with  $T_c \sim 5$  K) other than that of the bulk metal which may be a result of small amounts of impurities during deposition [Bond, W., 1965]. This is not readily controllable and makes it very difficult to reproduce single layer systems from these materials. In contrast, a reduction in  $T_c$  has been observed in films of some elements. Nb and Ta have significant reductions in  $T_c$  over the corresponding bulk values while Cr, Rh, and Pt films show no superconductivity above 300 mK [Bond, W., 1965].

#### 6.4.2.1 Tungsten

Since bulk tungsten has a particularly low transition temperature ( $T_c \sim 15$  mK), it has been the subject of much research as a transition edge sensor [Colling, P., 1996]. Bulk tungsten is in the  $\alpha$  body centred cubic (bcc) phase whereas thin films of W are typically deposited in the  $\beta$  (A15) phase, which is a suboxide (an oxide containing less than the normal proportion of oxygen) of the form W<sub>3</sub>O. This typically increases  $T_c$  from 15 mK to 0.6 K - 4 K. Since oxygen is thought to stabilise the crystal phase, the group at Stanford University etch to remove the oxide before deposition by sputtering [Irwin, K., 1995b]. They also heat the silicon substrate but have had best results by using d.c. sputtering with -200 V substrate voltage. With this technique they obtained reproducible transition temperatures of 70 mK with widths of 0.7 mK [Irwin, K., 1995b]. Similarly, tungsten films are successfully fabricated at the Max-Planck-Institut fur Physik, Munich for calorimeters developed by Oxford University for the Cryogenic Rare Event Search with Superconducting Thermometers (CRESST) dark matter search experiment [Bruckmayer, M., 1997].

#### 6.4.2.2 Molybdenum

Researchers at Goddard Space Flight Center investigated Mo layers as part of their Mo/Au programme. They deposited 50 nm thick Mo layers on SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> substrates at 400 °C and 800 °C [Finkbeiner, F., 1998]. The 800 °C samples showed sharper transitions and were lower stress films. These films had relatively high transition temperatures of 700 mK - 710 mK on SiO<sub>2</sub> and between 580 mK - 590 mK on Si<sub>3</sub>N<sub>4</sub>. This is too high for a low noise detector (hence the development of Mo/Au bilayers), but the transitions had widths of 3-5 mK and were reported to be reproducible.

# 6.4.2.3 Titanium

SRON deposited 8 Ti layers by evaporation and sputtering as part of their Ti/Au programme. The layer thickness ranged from 16 - 49 nm with transition temperatures ranging from 155 mK - 475 mK. However, the transition temperature is not a clear function of thickness and so reproducing a given  $T_c$  is difficult, in contrast to the manner in which a Ti/Au bilayer may be tuned to a target transition temperature.

#### 6.4.2.4 Iridium

A very promising candidate material for a TES is iridium [Fukuda, D., 1999]. As part of the Leicester detector programme, Oxford Instruments have demonstrated that a thin film (100 – 200 nm) of iridium

can be fabricated with excellent superconducting transition properties and a tuneable transition temperature in the required range (60 - 150 mK). Figure 6.2 shows an example of one of our iridium transition curves.



**Figure 6.2:** R(T) measurement of a test Ir TES from Oxford Instruments showing normalised resistance as a function of temperature across the transition.

The electrical properties of iridium-based devices are very stable. We stored 6 silicon wafers, each containing 15 Ir test TES resistors, for 17 months at room temperature and normal atmospheric conditions and saw no change in room temperature resistance in any sample (see table 6.2). Therefore, we conclude that post-processing oxidation does not significantly degrade iridium TESs.

#### 6.5 Auger Electron Spectroscopy (AES)

In order to determine the extent of interdiffusion across metallic interfaces relevant to TES design, we conducted destructive Auger Electron Spectroscopy (AES) tests on a number of samples in collaboration with the Institute of Surface Science and Technology at the University of Loughborough.

### 6.5.1 AES Theory

The surface of the sample is irradiated with a beam of 3-10 keV electrons which excite core level electrons in the sample atoms. The core level electrons are emitted from the parent which relaxes by emission of characteristic Auger electrons [Krause, M., 1975]. Only Auger electrons emitted from the surface have a significant probability of escape, resulting in an analysis depth of a few atomic layers ( $\sim$ 2 nm) - making AES a very sensitive surface identification technique.

For our depth profiling experiments we used alternating periods of AES measurement and sputtering with an inert ion beam (argon). The depth is not measured directly and so approximate depths only are given in the plots. Depth calibration is derived from the sputtering time and the rate of etching for each material (e.g. the sputter rate for gold is about three times that for titanium). A typical rate of etch is  $\sim 1$  nm in 30 seconds.

#### **6.5.2 AES Equipment Description**

Our experiments were conducted on a JEOL 7100 Auger electron spectrometer, the first of its kind in the UK. This spectrometer takes fully automated depth profiles. The sample is rotated during measurement to ensure that the sputtering crater surface is smooth and uniform (~1 mm in diameter). Without this feature, accurate depth profiles cannot be obtained. The analysis diameter is approximately 60 nm and the detection limit is about 0.1 % for all elements except H and He.

#### 6.5.3 AES Ti/Au Bilayer Result

The depth profile of a Ti/Au bilayer TES fabricated by SRON in August 1998 was measured in April 2000, after storage at normal room temperature, pressure and humidity conditions. This bilayer consisted of a 22 nm gold layer deposited over a 26 nm titanium layer on a silicon nitride substrate.



**Figure 6.3a**: AES result for a Ti/Au TES. The profiles for Ti and Au only are shown. The total etch time was 250 s and the depth scale is derived from this. The depth scale is approximate because Ti as oxide sputters at a different rate from Ti as metal, and in the sample it appears to be a mixture of the two.



**Figure 6.3b:** AES result for Ti/Au bilayer showing the profiles for oxygen, silicon and carbon. An approximate position for the Ti/Au interface is shown, as deduced from figure 6.2a.

Figure 6.3a/b shows that there has been significant interdiffusion across the Ti/Au interface, which was originally at a depth of 22 nm. There is also evidence of contamination on the surface of the sample, most notably carbon. Surface contaminants appear to have diffused (probably by grain boundary diffusion) into the sample and degraded the gold layer. This accounts for the relatively small amplitude of the gold peak (~50%). The grain size of the gold is 10 - 150 nm, but the titanium is typically smaller at 10 - 30 nm for a deposition temperature of ~150 °C [Bruijn, M., 1999]. Under these conditions, Ti compounds are expected to fill the grain boundaries around the gold grains during deposition. In such a thin gold layer (22 nm thick), the grain boundaries may extend from the metal interface to close to the surface. Therefore, the occurrence of a titanium signal in the gold layer can be accounted for by atomic diffusion during deposition. To confirm this hypothesis, AES studies should be conducted on samples of different ages. Such investigations are planned for Ti/Au and a range of other material combinations in the forthcoming year.

The oxygen contribution follows a characteristic profile: a continuous presence peaking at the surface and towards the silicon interface. This profile is seen in all depth measurements, irrespective of the materials involved. In this example a strong oxygen signal is also seen across the Ti/Au interface. This was commonly seen in highly diffusive systems such as Al/Ag (figure 6.5) and is indicative of a disrupted interface. Oxygen, and other gas atoms such as C and N, often diffuse as interstitial atoms in bcc metals such as W and Nb, and in hexagonal close packed (hcp) metals such as Ti. These small gas atoms readily fit into the interstices in the lattice and hence have negligible formation energy ( $S_f$ and  $H_f$  in equations (6-3) and (6-4)). For this reason, diffusion by impurity interstitials may be very fast. This process may account for the unusually strong C and O signals throughout the Ti layer. However, in this instance it should be noted that the presence of oxygen in the titanium layer most probably originates from the vacuum during deposition since titanium react so rapidly with any residual oxygen.

In general, interstitial impurities are difficult to exclude but the presence of carbon is not expected to directly compromise the performance of the TES. However, interstitial oxygen may either act as a barrier layer between the normal and superconducting layers, or accelerate oxidation of materials such as aluminium and thereby degrade the interface with the normal metal and compromise the proximity effect.

The transition temperature of 150 mK for this sample was measured at SRON shortly after fabrication in 1998. We repeated this measurement in our laboratory in April 1999 and showed that the transition was at 155 mK, and so had not changed significantly. Small differences of this order may be expected in the thermometry between the SRON and Leicester cryogenic facilities. Such consistency in  $T_c$  over time indicates that interdiffusion to the extent seen in this sample need not compromise the transition characteristics. We hypothesise that this applies to all immiscible material combinations, such as Mo/Au and Ta/Ag, where interdiffusion is not followed by the formation of new phases or intermetallic compounds. However, the transition characteristics are often radically altered in more reactive combinations, such as Al/Ag, where interdiffusion precedes the growth of new phases.

# 6.5.4 AES Ag/Al/Ag Trilayer Result

As described in section 6.4.1.1, we designed a Ag/Al/Ag trilayer to encapsulate the aluminium layer and protect it from the corrosive effects of chemicals used in processing. In addition to the reduced oxidation of the aluminium layer, a multilayer approach enables the normal resistance value to be reduced, for given  $T_c$  and TES dimensions. This is important since a TES with a lower normal resistance is expected to have a smaller thermal fluctuation noise spectral density, and hence improved energy resolution in the thermal fluctuation noise limit [Hoevers, H., 1999a].



**Figure 6.4**: Normalised resistance as a function of temperature for two 20/30/20 nm Ag/Al/Ag trilayers. Plotted in the broken lines are the corresponding  $\alpha \left(\frac{T}{R}\frac{dR}{dT}\right)$  values (right hand scale).

Figure 6.4 shows the transitions for two 20/30/20 nm Ag/Al/Ag trilayers made for us by Oxford Instruments. The superconducting properties are ideal for a TES detector. The transitions are sharp (transition width,  $\Delta T < 2$  mK), show good symmetry and high values of  $\alpha$  (~ 400). However, dramatic changes with time to both the normal resistance at room temperature and the  $T_c$  of these films indicated inherent instability (sections 6.6 and 6.7). TES processing repeated under different conditions to determine whether this instability resulted from a manufacturing process such as chemical etching proved negative. However, AES testing showed that the observed changes in the electrical properties were due to a combination of penetration of oxygen through the surface Ag layer and interdiffusion across the metal interfaces, figure 6.5.



**Figure 6.5**: Element profiles from an Auger Electron Spectroscopy measurement of a Ag/Al/Ag trilayer, showing distribution of Ag throughout the Al layer and oxidation at the first Ag/Al interface. The sample was 6 months old, and had been stored at room temperature.

The depth profile of the Ag/Al/Ag trilayer shows that there has been significant degradation during the 6 months between fabrication and testing. The lack of a distinct central aluminium layer clearly indicates that extensive interdiffusion had occurred. This is a consequence of the miscibility of these materials, which in turn accelerates the formation of new aluminium phases. Phase diagrams [Moffatt, W., 1986] reveal that aluminium systems are generally complex with many intermetallic compounds. Aluminium is soluble in silver and long range ordering can occur in the alloy [Markwitz, A. et al., 1998]. Such interdiffusion effects are thermally activated and so may be controlled by storing the thermometers at low temperatures (~77 K would effectively eliminate interdiffusion), but this is not an ideal practical solution. Furthermore, note the similarity in the oxygen profile with that in figure 6.3. An oxygen peak at the upper Ag/Al interface is evidence of oxygen penetration of the surface Ag layer, again most probably by grain boundary diffusion. This has resulted in a 10 nm thick oxide on the top side of the aluminium layer and is an important mechanism by which these trilayers degrade.

#### 6.5.5 AES Cu/Nb/Cu Trilayer Result

Although niobium has the highest transition temperature of any element (9.2 K), significant reductions in  $T_c$  are seen in thin Nb films [Bond, W., 1965]. The Cu/Nb system is immiscible [Moffatt, W., 1986] and so is a suitable candidate for investigation, but is not currently considered as a basis for a

practical TES due to the high transition temperatures. As part of the UK Cryogenic X-ray Spectrometer Programme (XCSP), Cambridge University Materials Department deposited a series of Nb/Cu bilayers, trilayers and multilayers using their sputtering chamber (which deposits films of higher purity than their evaporator). Their results show that the transition temperature varies inversely with number of layers, even if the total thickness of Nb and Cu is constant (figure 6.6). In each case, 70 nm of copper was used and the transition temperature,  $T_c$  measured as a function of niobium thickness and number of layers.



**Figure 6.6**: Transition temperature as a function of Nb layer thickness for Nb/Cu bilayers (open circles), trilayers (closed circles) and multilayers (crosses) [Burnell, G., 2000]. In each case, the copper layer is of thickness 70 nm.

From figure 6.6 it is clear that fabricating a Nb/Cu multilayer with a  $T_c$  in the required range for a TES (80-150 mK) is difficult. However, the interdiffusion dynamics were still of interest, particularly since in the future we may use films of copper in conjunction with bismuth – a similarly immiscible system (Chapter 9). Results from the Auger electron spectroscopy profile of a 35/18/35 nm trilayer are shown in figure 6.7. This plot includes data (solid lines) taken after our colleagues in Loughborough heated the wafer to 100 °C for 24 hours to enhance diffusion effects. By inspection of figure 6.7, it is clear that the Cu/Nb boundary interfaces are well defined and were not changed significantly by the period of elevated temperature. The most notable difference is the redistribution of oxygen through the system. The interface with the substrate was largely unaffected by increased temperature, but as expected, heating caused increased oxidation of the outer copper surface. Moreover, the oxygen in the niobium layer (probably a residual from the deposition process) migrated to interfaces with the copper. This is shown more clearly in figure 6.8.



**Figure 6.7**: AES profile of a 35/18/35 nm Cu/Nb/Cu trilayer. The broken lines show the composition (12 months after fabrication) before the sample was heated to 100 °C for 24 hours. The subsequent analysis is represented by the solid lines.



**Figure 6.8**: Expanded view of the region of interest (30 - 60 nm) for the migration of oxygen as a result of heating the sample to 100 °C for 24 hours.

Heating has induced a smoothing of the oxygen profile centred on the Cu/Nb interfaces. The quantity of oxygen is small (< 5% of the total composition) but the sensitivity of the AES technique is such  $(\pm 0.1 \%)$  that we have confidence in the results. This is a significant result because redistribution of oxygen through a system may change the transition temperature by stimulating oxidation at the interface and acting as a barrier between the two proximity layers. The implications of this process may have serious consequences for other material combinations, and a series of experiments are planned for later in 2001 to analyse the oxygen distribution as a function of temperature/time for all relevant material interfaces on our detectors (Nb/Au, Nb/Ag, Ir/Au, Ir/Nb – see Chapter 5).

#### 6.6 Changes in Resistance at Room Temperature as a Function of Time

Tracking changes in resistance at 300 K is a simple method of monitoring film degradation. Further work is required to determine the exact nature of the correlation between changes in room temperature resistance and changes in transition temperature, but our experiments with Al/Ag indicate that these two properties are related. Figure 6.9 shows the typical change in normal resistance at room temperature of a Ag/Al/Ag trilayer over a period of about 10 days. The data set is limited, but is well described by a linear fit. In contrast, similar tests conducted on Ti/Au systems show no significant change in resistance over periods of months. Therefore, resistance changes at 300 K are a further measure of degradation of Al/Ag films, accounted for by diffusion related processes.



Figure 6.9: Resistance of a Ag/Al/Ag trilayer at room temperature as a function of time stored in the laboratory.

In distinct contrast, no significant changes in room temperature resistance over a period of 17 months have been observed in our Ir TESs, see table 6.2.

Ir Sample	Mean Resistance (k $\Omega$ )	Mean Resistance (k $\Omega$ )	Mean Resistance (k $\Omega$ )
	October 1999	June 2000	March 2001
1	1.99	1.99	2.00
2	1.94	1.92	1.96
3	0.86	0.87	0.87
4	0.85	0.85	0.84
5	1.02	1.02	1.01
6	2.07	2.07	2.08

**Table 6.2:** Room temperature resistance of Ir samples over a 17 month period. Each sample contains 15 resistors and the mean resistance value has been taken. No significant changes have been observed.

# 6.7 Changes in Transition Temperature as a Function of Time

# 6.7.1 Ag/Al/Ag

Figure 6.10 shows the change in transition temperature,  $T_c$  over time for two Ag/Al/Ag trilayers. The measurements were separated by several days of storage at room temperature in the laboratory. The profile of the transition does not change significantly over this period, but there is a clear reduction in the transition temperature. In general, this is consistent with a depletion of the superconductor layer, aluminium in this instance. The possible depletion mechanisms are interdiffusion of the two metals across the interface and oxidation of the aluminium. Auger spectroscopy measurements indicate that both processes occur (section 6.5.4), but the short timescales (~ days at room temperature) for degradation of these samples suggests that oxidation processes are dominant.



Figure 6.10: The change in transition temperature for two Ag/Al/Ag trilayers over successive measurements taken a few days apart. Oxygen diffusion through the Ag grain boundaries leads to oxidation of the Al layer and a decrease in Tc. Resistance is quoted in arbitrary units. No transition above 300 mK was seen for film 1 during the third test.

#### 6.8 Conclusion

The ageing of TES materials and reliability of detectors are key issues in the development of an astronomical X-ray spectrometer. Auger analysis and resistance measurements on Al/Ag exposed the weakness of this combination and lead us to investigate other materials, such as Ti/Au and Ir. Although apparently stable and possessing a simple phase diagram [Moffatt, W., 1986] Ti/Au systems do show diffusion across the interface. Auger analysis has also been used to successfully show transport of oxygen through Nb/Cu trilayers as a result of exposure to elevated temperatures and further work should be conducted to determine the likely effect on resistance of this structure, and other material combinations, over time.

The question of which is the best material for a TES remains unanswered, and is probably application specific. Currently, for an X-ray spectrometer, we consider iridium to be the most promising candidate material. Iridium yields sharp and stable transitions and, as a monolayer, has the advantage of offering higher bias currents (and therefore higher potential countrate) than equivalent bilayer configurations. A separate experiment has been conducted to investigate the superconducting properties of iridium in detail, and this is described in Chapter 7.

# **Chapter 7**

# The superconducting properties of iridium

# 7.1 Iridium as a TES Material

As described in section 2.2.2.2, a monolayer TES has several advantages over a multilayer approach. The choice of superconductor is, however, limited because a low critical temperature is required. Possible candidates include tungsten (bulk  $T_c \sim 15$  mK) and hafnium (bulk  $T_c \sim 128$  mK), but the literature indicates that generating consistent transitions in the range 80 – 120 mK in thin films is difficult for these materials [Bruckmayer, M., 1997; Kraft, S., 1998a]. Iridium is another candidate material with low bulk  $T_c$  (112 mK) and good chemical stability. This Chapter describes a series of experiments to measure the superconducting properties of thin film Ir and assess its use in a TES detector. Little had been published on the use of iridium in TES technology when our investigation into new materials commenced [Frank, M., 1994; Nagel, U., 1994; von Kienlin, A., 1998]. Results published subsequently are consistent with ours [Fukuda, D., 2000].

#### 7.2 Iridium Material Description

Iridium is a transition metal with an atomic number of 77, is very hard and brittle and the most corrosion resistant metal known. The superconducting properties of bulk iridium were first reported by Hein et al. [1962]. Since then, the transition temperature for pure bulk samples has been thoroughly investigated [Andres, K., 1968, Wejgaard, W., 1969] with convergence of results at 0.1125 K  $\pm$  0.0005 K [Gubser, D., 1973]. The reported transition temperatures for thin films depart considerably from the bulk value, depending on the deposition parameters and substrate used: from 91 mK [Nagel et al., 1994] to 326 mK [Fukuda, D., 2000].

#### 7.2.1 Electrical Resistivity of Bulk Iridium

Matthiessen's Rule states that the total resistivity,  $\rho_T$ , of a metal is made up of electron scattering processes that add independently [Ohring, M., 1992]:

$$\rho_T = \rho_{Th} + \rho_D + \rho_I \tag{7-1}$$

Where  $\rho_{Th}$ ,  $\rho_D$  and  $\rho_I$  are the resistivities attributable to thermal, defect and impurity effects.

All of these components arise due to disruptions in the periodicity of the atomic lattice. In the case of thermal resistivity, the usual scattering mechanism is electron – phonon scattering. This scales linearly with temperature and is dominant at room temperature (for pure films). Electron scattering off defects, vacancies or impurities in the lattice is independent of temperature and is usually important at low temperatures only.

One characteristic of the transition metals is their high room temperature resistivity compared to alkali and noble metals (5.5  $\mu\Omega$ cm for Ir at room temperature [Handbook of Chemistry and Physics, 1989]). Mott [1936] explained this in terms of overlapping d and s band electrons, where d and s represent different angular momentum states. In transition metals, d electrons occupy a narrow energy band with a high density of states. This implies a small value for  $\frac{dE}{dk}$ , where E is the electron energy and k is the wave vector. The velocity of an electron is given by [Rosenberg, H, 1963]:

$$v = \left(\frac{2\pi}{h}\right) \frac{dE}{dk}$$
(7-2)

Therefore, the velocity of the d band electrons is relatively small and will not contribute much to the electric current. Another consequence of the high density of states for d band electrons is that there is a strong probability that the s electrons (which have a higher velocity and therefore carry most of the current) will be scattered into vacant d states. This s-d interaction is additional to the phonon interactions important in all metals and is used to explain the high resistivity of transition metals at room temperature.

#### 7.2.2 Electrical Resistivity of Thin Film Iridium

In general, electrical resistivity is increased when a bulk metal is reduced to a thin film. The electrical properties of metals are highly dependent on the film growth conditions, since deposition parameters (such as substrate temperature, rate of deposition and purity of source) determine film parameters such as crystal perfection, grain morphology, defect and void density and quality of the interface with the substrate. In addition, as the thickness of the film, t, is reduced, there comes a point where the thickness is less than the effective electron mean free path length,  $\lambda_{eff}$  (~ 100 nm) and scattering from the film surfaces becomes important and acts to increase film resistivity.

# 7.2.2.1 Grain Boundary Scattering

Grain boundary scattering,  $\rho_{GB}$  is a contributor to the term  $\rho_D$  in equation (7-1). Scattering of electrons by grain boundaries is an important effect for thin films where the crystallite size is less than the electron mean free path. Grain diameter is determined by deposition temperature (Chapter 5), therefore resistivity due to grain boundary scattering is also a function of deposition temperature. If it is assumed that the average grain size is equal to the film thickness and that the only contribution to electron scattering is from the boundaries with normals in the film plane, then  $\rho_{GB}$  is given by [Mayadas, A., 1970]:

$$\rho_{GB} = \frac{\rho_0}{3\left[\frac{1}{3} - \frac{1}{2}\beta + \beta^2 - \beta^3 \ln\left(1 + \frac{1}{\beta}\right)\right]}$$
(7-3)

Where  $\rho_0$  is the bulk resistivity of the metal and  $\beta$  is a parameter related to the grain boundary scattering relaxation time:

$$\beta = \left(\frac{\lambda_{\rm o}}{D}\right) \left(\frac{R}{1-R}\right) \tag{7-4}$$

Here, D is the average grain diameter (equal to t) and R represents the probability of backscatter upon reflection at a grain boundary. For R = 1, electrons are reflected along their original trajectory and the resistivity becomes infinite. A value of R = 0.2 has been assumed in subsequent calculations (R = 0.17 for Al and R = 0.24 for Cu [Ohring, M., 1992]).

The electron mean free path of the bulk material,  $\lambda_0$ , is given by [Reale, C., 1974]:

$$\lambda_0 = \left(\frac{h}{2e^2\rho_0}\right) \left(\frac{3}{\pi N_s^2}\right)^{\frac{1}{3}}$$
(7-5)

Where *h* is Planck's constant, *e* is the charge on an electron,  $\rho_0$  is the bulk resistivity and  $N_s$  is the carrier concentration (*s* band electrons only). For  $N_s = 5 \times 10^{28} \text{ m}^{-3}$  [American Institute of Physics Handbook, 1972] and  $\rho_0 = 5.5 \,\mu\Omega$ cm [Handbook of Chemistry and Physics, 1989],  $\lambda_0 = 17 \text{ nm}$ .

Substitution of equation (7-4) into (7-3), with  $\lambda_0 = 17$  nm and D = t = 100 nm (a typical thickness for our experimental Ir films), yields:

$$\frac{\rho_{GB}}{\rho_0} = 1.06$$

#### 7.2.2.2 Thin Film Surface Scattering

Electron interaction with the film surfaces is important when interaction with the film surface stops the electron from executing its full mean-free path. If the scattering is diffusive (this is dependent on the nature of the film surface) then an additional component to the resistivity arises. If the scattering is totally specular there is no dependence of the resistivity on film thickness. Work by Thomson [1901] was developed by Fuchs [1938] and Sondheimer [1951] to include the quantum behaviour of free electrons and the statistical distribution of mean free paths. For thick films,  $\frac{t}{\lambda_0} \ge 1$ , their model predicts a film resistivity due to surface scattering,  $\rho_s$ , where:

$$\rho_{s} \approx \frac{\rho_{0}}{1 - \frac{3\lambda_{0}}{8t} \left(1 - P\right)}$$

$$(7-7)$$

where P (0 < P < 1) is the proportion of scattering events that are specular. If purely diffusive scattering is assumed, P = 0 and taking the same values for  $\lambda_0$  and t as above yields:

$$\frac{\rho_s}{\rho_0} \approx 1.06 \tag{7-8}$$

The dependence of grain boundary and surface scattering on film thickness is shown in figure 7.1. Purely diffusive scattering has been assumed (P = 0). The plots are terminated for t < 20 nm because the equations describing the curves are valid for thick films only ( $\frac{t}{\lambda_0} \ge 1$ ).

By comparison of equations (7-6) and (7-8) and by inspection of figure 7.1, it is clear that, from resistivity measurements alone, it is difficult to determine whether any change in Ir resistivity as a function of film thickness is due to grain boundary or film surface scattering. In the next section, we consider how temperature measurements at constant film thickness can be used to obtain information on defect and impurity-related resistivities.



**Figure 7.1**: Effects of surface scattering and grain boundary scattering on Ir thin film resistivity as a function of film thickness, t. Our experimental films were either 100 nm or 200 nm thick; in this regime both effects are small and show similar dependences on t.

# 7.2.3 The Residual Resistivity Ratio (RRR)

Differences in the ways in which thermal, impurity and defect contributions to resistivity scale with temperature may be used to assess the perfection of a thin film. The Residual Resistivity Ratio (*RRR*) is defined by [Gubser, D., 1973]:

$$RRR = \frac{\rho_T(300K)}{\rho_T(4.2K)}$$
(7-9)

Since thermal processes dominate at 300 K but are frozen out at 4.2 K, the *RRR* may be approximated to:

$$RRR \sim \frac{\rho_{Th}(300K)}{\rho_D(4.2K) + \rho_I(4.2K)}$$
(7-10)

From equation (7-10) it is clear that pure, defect-free films will yield high ratios. Values for sputtered films range from  $RRR \sim 1$  to 2000 [Gubser, D., 1973]. For extremely pure films, resistivity measurements remain the only technique available for determining purity because chemical methods fail due to their limited sensitivity. Purity is important in our application because the transition

temperature may deviate from the bulk value in the presence of contaminants. Gubser et al. [1973] suggest that even small concentrations of impurities in bulk material may lead to anisotropy in the energy gap that strongly affect the  $T_c$  of the sample. Iridium is also particularly susceptible to ferromagnetic contamination in either the Ir sample or the vacuum system used for film preparation, and this may suppress the transition temperature [Nagel, U., 1994]. In addition, pure Ir is a type I superconductor whereas impure samples may exhibit type II characteristics including considerable trapped flux in zero applied field [Gubser, D., 1973]. This may contribute to the excess detector noise observed by SRON, Utrecht (section 2.7.4). The purity of our iridium target was 99.8%, as certified by the Ir manufacturer, Kurt J. Lesker Co. Ltd (16 Ivyhouse Lane, Hastings, E. Sussex). The most common impurities are Rh (200 ppm), Pb (30 ppm), Fe (30 ppm), Ca (30 ppm), Si (20 ppm), Pd (20 ppm) and Na (20 ppm). The extent to which impurities have affected our results could only be ascertained by further testing with a target of different purity, but the data indicate that (at this level of purity) a correlation exists between transition properties of thin films and substrate deposition temperature (section 7.5).

#### 7.3 Deposition Parameters

Thin iridium films may be deposited by electron beam evaporation [Wagendristel, A., 1994], or, due to the relatively high melting point of the metal (2739 K), sputtering [Heavens, O., 1970]. In general, the condensing atoms of sputtered films have relatively high energy (2-30 eV [Ohring, M., 1992]) so such films typically consist of smaller grains with good substrate adhesion. Sputtered films may incorporate gas atoms from the carrier gas (often argon) whereas evaporated films may be contaminated by residual gas in the vacuum. There is also evidence that the presence of electrons or ions in the condensing beam may have a marked effect on the film properties [Heavens, O., 1970]. Thus, even under apparently identical deposition conditions, films often show differences in structure and electrical properties. An alternative fabrication method is to polish a bulk sample down to the required thickness [von Kienlin, A., 1998], but it is difficult to fabricate a sufficiently small volume by this means with the low specific heat necessary for high energy resolution detectors.

Our Ir samples were prepared at Oxford Instruments, Cambridge using their Chemical Vapour Transport (CVT) UHV magnetron sputtering system. The deposition rate on to the silicon nitride substrate surface was 10nm/min and the vacuum pressure prior to deposition, about  $10^{-9}$  mbar. Film thickness was measured with a Taylor Hobson Talystep. This consists of a stylus that is brought into contact (post-facto) with the film and scanned across the surface. Steps in the profile at the edge of the film may be measured to an accuracy of 2 %, without significant damage to the film surface. The transition temperature,  $T_c$  is expected to vary as a function of substrate deposition temperature. Several deposition runs were made at different temperatures, ranging from 350 – 800 °C, to characterise this functionality. In general, higher temperatures produce films of lower resistivity and

 $T_c$ , with the increased risk of impurity diffusion into the film from the substrate, particularly for substrates with low melting points such as silicon (1687 K).

# 7.4 The Iridium Superconducting Transitions Experiment

Prior to the production of Ir sensors as part of the Leicester detector programme, we commissioned Oxford Instruments to fabricate a series of low internal mechanical stress (i.e. neither under tension nor compression with respect to the substrate) Ir test structures (see figure 7.2). These consisted of Ir meander tracks, deposited using an existing lithography mask set, terminated by bond pads. Six deposition runs were conducted. Four different deposition temperatures were used, and the film thickness was also varied for one of these temperatures (see table 7.1). Two of the chips were manufactured under identical conditions, but on different dates. This enabled us to differentiate between (a) transition temperature differences induced by irreproducibility and (b) deliberate changes in the deposition parameters. Reproducibility is a key factor for detector development. There were 15 resistors on each chip. Measurements were made on at least two resistors from different positions on each chip to investigate any systematic variations in critical temperature across the chip surface, giving 12 resistor measurements in total.

I conducted the test in Cambridge using the liquid helium dilution refrigerator (base temperature ~ 15 mK) at Oxford Instruments. This system has additional wiring compared to the Leicester facility and could support the simultaneous measurement of all 6 chips in a single cooldown. Resistance measurements were made using a four-point technique through an a.c. resistance bridge. An additional bridge was used to monitor the thermometers. The thermometry consisted of a 2200  $\Omega$  RuOx resistor thermometer on the mixing chamber (which is only accurate for T > 100 mK), and two other 2200  $\Omega$  RuOx resistor thermometers (T > 20 mK), one fully calibrated. The uncalibrated RuOx resistor was mounted on the mixing chamber whereas the calibrated thermometer was mounted at the base of the sample stage. The offset between these two readings was constant over a range of temperatures; suggesting there was not a significant thermal gradient down the sample stage.





The superconducting-to-normal transitions were typically very sharp and for accurate data logging (recorded from multi-channel AVS resistance bridges) it was necessary to simultaneously record rapid changes in the resistance and thermometry data. For practical reasons this was best achieved using the uncalibrated RuOx thermometer. Throughout the measurement run, simultaneous measurements of both thermometers at a range of temperatures were made, and the resultant relation is best described by a linear fit with a systematic offset, see figure 7.3.

To determine the temperature from measurements taken by the uncalibrated thermometer, the uncalibrated values were first corrected by the offset shown in figure 7.3. These new values were compared to the best fit line of figure 7.4. This is a plot of the look-up table values for the calibrated thermometer. Errors in this calibration process are estimated to be sub-mK and insignificant when compared to other possible sources of error such as poor thermal contact between the sample stage and resistor substrate and self-heating of the resistors when in the normal state.



Figure 7.3: Correlation between the calibrated and uncalibrated RuOx thermometers. A linear fit to the data is assumed. The regression coefficient is  $R^2 = 0.9995$ .



Figure 7.4: Temperature calibration for the primary thermometer over the temperature range 40 - 160 mK.

The rate of change in temperature through the transition was varied for each sample from approximately 10 mK/min to 1 mK/20 mins but no corresponding change in transition width or position was observed. The temperature was controlled by use of a variable heater input on the mixing chamber. The transitions (figure 7.5a-f) are plotted from data acquired at the slowest possible sweep rate so thermal time lags between the sample and thermometer may be considered negligible.

# 7.4.1 Results and Discussion

#### 7.4.1.1 Resistivity Measurements to 4.2 K

The resistivity at 300 K and the *RRR* were measured for each of the 6 samples. The results are shown in table 7.1. Sample numbers 1 and 3 were grown at the same deposition temperature (400 °C, controlled by Oxford Instruments) but to a different layer depth. Comparison of the resistivity of samples 1 and 3 shows that, as expected, the thinner layer is of higher resistivity. As stated in section 7.2.2.2, this may be due to either enhanced grain boundary or surface scattering. However, since the deposition temperature (which is the most important parameter in determining mean grain size) is a constant, the increase of 12 % in the room temperature resistivity may be attributed primarily to enhanced surface scattering. Sample 5 was deposited at the highest temperature (800 °C) and sample 6 at the lowest temperature (350 °C). These films were grown to the same thickness and so the resistivity characteristics of these films may be attributed to differences in grain structure.

The room temperature resistivity values were reproducible to an accuracy of approximately 10 %. The *RRR* was reproducible to a similar accuracy. Our *RRR* values are similar to those obtained by other groups working with Ir films. Fukuda et al. [2000] report *RRR* values of 1.71 for a 40 nm film sputtered at room temperature onto a  $Si_3N_4$  substrate, and 1.5 when deposited onto a Si substrate under the same conditions. Frank et al. [1994] quote an *RRR* value of 2.5 for a 400 nm film evaporated onto a Si substrate at 520 °C. By comparison with these published results, we may conclude that our sputtered films are of equivalent purity and structure to evaporated films and therefore it may be concluded that no significant quantities of carrier gas are incorporated into our films.

Sample number	Film thickness (nm)	Resistivity (300 K) (μΩcm)	<i>RRR</i> (4.2 K)
1	$100 \pm 5$	13.7±0.7	$1.74 \pm 0.01$
2	100 ± 5	14.3±0.7	$1.74\pm0.01$
3	$200 \pm 10$	12.2±0.6	1.93 ± 0.01
4	$200 \pm 10$	11.4±0.6	1.94 ± 0.01
5	100 ± 5	6.8±0.4	$3.70\pm0.01$
6	$100 \pm 5$	$14.5 \pm 0.7$	$1.71 \pm 0.01$

**Table 7.1**: Resistivity data for Ir test resistors. The degree of shading corresponds to deposition temperature (350 – 800 °C, dark shading implies high temperature). Sample numbers 1 and 2 were grown to the same specifications but on different dates, as were samples 3 and 4. Sample 5 was deposited as a low  $T_c$  film (~ 52 mK), and sample 6 was a high  $T_c$  film (~ 157 mK). This follows from the expected relation of high temperature deposition leading to low  $T_c$ , and low temperature deposition leading to high  $T_c$ .

# 7.4.1.2 Transition Data

The following plots show examples of transitions for each of the six Ir samples. In each case, two resistors have been measured on each chip (blue curves and green curves). The data were taken both with increasing and decreasing temperature to expose any self-heating effects. No significant hysteresis can be seen between the data sets of increasing temperature (dark blue/green) and decreasing temperature (light blue/green). All graphs are drawn to the same temperature scale for direct comparison of  $\Delta T$ . Small-scale structure in the transitions is most likely the result of measurement errors due to the rapid change of resistance with temperature.



Figure 7.5a: superconducting transitions for sample 1.



Figure 7.5b: superconducting transitions for sample 2.



Figure 7.5c: superconducting transitions for sample 3.



Figure 7.5d: superconducting transitions for sample 4.



Figure 7.5e: superconducting transitions for sample 5.



Figure 7.5f: superconducting transitions for sample 6.

For each sample, the transition with most data is plotted in figure 7.6. The transitions have been normalised for ease of comparison of transition widths. This is justified since the normal state resistance of an actual TES device will be of the order 1  $\Omega$ . The test resistors have much higher resistances due to their long meander length. Therefore, the magnitude of the normal state resistance of the test resistors at the transition is not relevant when comparing transition profiles.



**Figure 7.6**: Comparison of normalised iridium transitions. The transition temperatures occur in the correct order expected from the deposition parameters.

The transitions occur in the expected order with close grouping of samples #1 and #2 (light/dark green) and TES #3 and #4 (orange/red). These resistor pairs were deposited under the same conditions but on different dates. The extreme  $T_c$  samples, #5 and #6 show transitions close to their predicted temperatures (60 mK and 140 mK, respectively). Predictions were based on empirical data of previous results. Figure 7.7 shows the comparison between expected and measured transition temperatures. This demonstrate a fair degree of control over the process, but the elevated measured transition temperatures for samples 1 - 4 (100 < Estimated  $T_c < 110$  mK) remain unexplained.





The  $\alpha$  values for these curves vary throughout the transitions, but an estimate for  $\alpha$  at the centre of each transition is given in table 7.2. Comparisons are made between our sputtered films, those of

Sample Number	Measured T <sub>c</sub> (mK)	α ((T/R)(dR/dT))
		(at midpoint of transition)
1	$148 \pm 1$	$300 \pm 100$
2	$155.5 \pm 0.5$	$350 \pm 100$
3	$135 \pm 1$	$100 \pm 50$
4	$139 \pm 0.5$	$500 \pm 100$
5	$52 \pm 2$	$20 \pm 10$
6	157 ± 1	$100 \pm 50$
Fukuda et al. (sputtered, 40 nm)	288.6	1273
Von Kienlin et al. (bulk, 40 µm)	109.5	164

Fukuda et al. [2000] (also on a  $Si_3N_4$  substrate) and the polished bulk device of von Kienlin et al. [1998].

Table 7.2: Transition properties for our iridium films in comparison with published results.

The errors in  $T_c$  and  $\alpha$  mainly arise from apparent differences between resistors on the same chip. It is possible that the measured differences result from differences in thermal contact with the substrate or thermal gradients across the chip due to self-heating effects, but the repeatable nature of this data set indicates that the differences are best attributed to variations in film structure. These  $\alpha$  values are also highly sensitive to errors in individual data points, which may have significant errors if the bath temperature is not stable. For this reason,  $\alpha$  is best calculated from a best fit line applied to the transition data.

# 7.5 Iridium Transition Temperature as a Function of Deposition Parameters

Of the few groups working with iridium films and publishing their results (Frank, M., [1994]; Nagel, U., [1994]; von Kienlin, A., [1998]; Fukuda et al. [2000]), none have provided a theoretical framework for prediction of transition temperature. These groups apparently use empirical data to guide their choice of deposition parameters. The published data set is limited but a pattern for sputtered films is clear: deposited films typically have a transition temperature *higher* than for bulk iridium, by a factor of up to 3. The classical expectation is for the transition to be suppressed due to enhanced scattering and reduced electron mean free path in a thin film. This is a similar mechanism to that responsible for the reduction in  $T_c$  by the addition of impurities to a bulk sample. However, thin films may, in general, show transitions deviating significantly from bulk values. Notable examples include bismuth, beryllium and tungsten. Bismuth is not known to be superconducting in the bulk state but thin films have transitions in the range 6.17 K – 6.6 K [Handbook of Chemistry and Physics, 1989]. Similarly,

beryllium has a low transition temperature of 26 mK for bulk samples but thin films show transitions as high as 8.4 K [American Institute of Physics Handbook, 1972]. Tungsten has been studied extensively and transitions for thin films in the range 0.6 K – 4.1 K have been reported [Irwin, K., 1995b], compared to a bulk value of 15.5 mK. In the case of tungsten, it is known that the modification in  $T_c$  is due to a change of phase upon deposition as a thin film. Bulk tungsten appears in the  $\alpha$  phase (bcc) but thin films usually deposit in the  $\beta$  phase (an A15 structure) [Bond, W., 1965]. It is possible that iridium also undergoes a similar phase change when deposited on a silicon or silicon nitride substrate. Evidence for this (assuming that a phase change is the mechanism responsible for modification of the transition temperature) is presented in figure 7.8, which shows a correlation between substrate deposition temperature and transition temperature for sputtered iridium films on silicon nitride substrates.



**Figure 7.8**: Transition temperature as a function of deposition temperature for iridium films on  $Si_3N_4$  substrate. The published result of Fukuda et al [2000] is plotted in addition to our data.

The data set plotted in figure 7.8 is limited and it is impossible to draw significant conclusions from a correlation defined by the single data points at the temperature extremes. However, initial indications support a hypothesis of a baseline transition temperature of  $\sim 300$  mK for thin film iridium, sputtered at room temperature, which is reduced by interaction with the silicon nitride substrate – an effect enhanced by higher deposition temperature and hence greater substrate interaction. The exact nature of the substrate interaction depends on the activation energies of various phases in the iridium-silicon system.



**Figure 7.9:** Partial iridium-silicon phase diagram, reproduced from Allevato [1993]. Open triangles indicate compounds determined by differential thermal analysis, closed triangles represent direct observation data points.

It is known that iridium silicides readily form when iridium is deposited onto single-crystal silicon substrates [Petersson, S., 1979]. This system is described by a complex phase diagram (figure 7.9). Petersson et al. identified three distinct, temperature dependent, phases: IrSi, IrSi<sub>1.75</sub> and IrSi<sub>3</sub> (although other authors report more than eight intermediate phases [Nicolet, M., 1983]). IrSi (orthorhombic) forms at temperatures between 400 °C and 600 °C, whereas IrSi<sub>1.75</sub> forms from 500 °C to 950 °C. Both of these phases have diffusion controlled growth with Si as the dominant diffusion species and have activation energies of the order of 2 eV. This is the activation energy for the diffusive process necessary for the growth of the IrSi layers. At 1000 °C, the IrSi<sub>3</sub> phase (hexagonal) forms by nucleation and growth processes, without any composition gradients through the film. The presumed mechanism for this is a rapid redistribution of silicon through the iridium layer leading to the formation of lower silicides and subsequent transformation into IrSi<sub>3</sub>, with an associated activation energy of approximately 3 eV. Other phases have been reported in the literature [Engström, I., 1970]. These include Ir<sub>4</sub>Si<sub>5</sub> (monoclinic), Ir<sub>3</sub>Si<sub>4</sub> (orthorhombic) and IrSi<sub>1.5</sub> (monoclinic), the last of which may be a misidentification of the IrSi<sub>1.75</sub> phase [Petersson, S., 1979].

Our deposition temperatures were all less than 1000 °C and so we can neglect the Si-rich IrSi<sub>3</sub> phase. The results of Petersson et al. [1979] suggest that it is also reasonable to assume that the IrSi and IrSi<sub>1.75</sub> phases are most important in our analysis.

# 7.6 Modelling the Ir Resistor as an Ir/IrSi bilayer

Many silicides have metallic properties, including high electrical conductivity, but iridium silicides are not known to be superconductors. If the reduction in  $T_c$  at elevated temperatures is due to the formation of iridium silicides, the system may be modelled as a proximity bilayer, with the silicide forming the normal metal component. The electrical resistivity values for iridium silicides are of order 5  $\mu\Omega$ m, or 10 % (or less) of the value for the silicon from which the silicide was formed [Allevato, C., 1993].

Various theories have been proposed to describe the proximity effect, reviewed by Gilabert, A. [1977], Banerjee, I., et al. [1982] and Chen, J. [1990]. Most share the concept of superconductivity by electron pair correlation, with an associated coherence length, in bulk superconductor, of order  $10^4 - 10^{-5}$  m. It follows that this correlation extends a similar distance into a normal metal in contact with the superconductor. Therefore, superconducting films of thickness less than the coherence length will have a transition temperature lowered by the presence of the normal metal. The effect is independent of the normal metal layer thickness, provided this is greater than the coherence length. In the case of a bilayer, this effect may be described by solving the Ginzburg-Landau equation [Ginzburg, V., 1950]. De Gennes and Geyon [1963] and Werthamer [1963] developed the theory in the limit of dirty films, where the electron mean free path,  $\lambda$  is less than the coherence length,  $\xi$ . The effective coherence length in thin film iridium is of order 200 – 740 nm, whereas the electron mean free path is typically 27 – 380 nm [Nagel, U., 1994]. Therefore, we compare our data to the de Gennes – Werthamer theory.

The coherence length in the superconductor,  $\xi_s$  is defined as [Hauser, J., 1964]:

$$\xi_s = \left(\frac{\hbar v_{Fs} \lambda_s}{6K_B T_c}\right)^{\frac{1}{2}}$$
(7-11)

Where  $v_{Fs}$  is the velocity of Fermi surface electrons in the superconducting layer and  $\lambda_s$  is the mean free path of electrons.  $T_c$  is the transition temperature of the bilayer,  $K_B$  is Boltzmann's constant, and  $\hbar$  is Planck's constant  $\frac{h}{2\pi}$ . Pippard [1960] established the relation between  $v_{Fs}\lambda_s$  and experimentally measurable quantities: the conductivity,  $\sigma$ , and the temperature coefficient of the electronic component of heat capacity,  $\gamma$ .

$$\upsilon_{F_s} \lambda_s = \left(\frac{\sigma}{\gamma}\right) \left(\frac{\pi K_B}{e}\right)^2 \tag{7-12}$$

Substitution of equation (7-12) into (7-11) yields:

$$\xi_s = \left(\frac{\pi^2 \hbar K_B \alpha}{6e^2 \gamma \rho_b}\right)^{\frac{1}{2}}$$
(7-13)

Where the resistivity,  $\rho$  has been replaced by  $\rho = \frac{\rho_b}{RRR}$ , with  $\rho_b$  = bulk resistivity, and  $\alpha$  is defined

by 
$$\alpha = \frac{RRR}{T_c}$$
. The coherence length is estimated assuming  $\gamma = 370 \text{ Jm}^{-3}\text{K}^{-2}$  [Nagel, U., 1994],

For our highest  $T_c$  sample (#6),  $\alpha = \frac{1.71}{0.157} = 10.89 \pm 0.09 \text{ K}^{-1}$  and  $\xi_s = 328 \pm 0.5 \text{ nm}$ .

For our lowest  $T_c$  sample (#5),  $\alpha = \frac{3.7}{0.052} = 71.2 \pm 2.7 \text{ K}^{-1}$  and  $\xi_s = 571 \pm 6 \text{ nm}$ .

These values may be compared to those for bulk pure iridium ( $\xi_s = 4400 \text{ nm}$  [Gubser, 1973]), and the values obtained by Nagel et al. [1994] for evaporated films ( $\xi_s = 200 - 740 \text{ nm}$ ). We may also compare the coherence lengths of our films with the electron mean free path for pure bulk iridium (16.7 nm) and for thin films as measured by Nagel et al. [1994] (27 – 380 nm) to justify our assumption of the dirty superconductor regime.

Asheda et al. [1992] have extended the theory for clean films ( $\lambda >> \xi$ ) and include the reflection coefficient of electrons at the normal – superconductor interface. Since our iridium films are neither clean nor likely to have a sharp interface with the silicide layer, the theory of Asheda et al. is not valid. We have applied the de Gennes - Werthamer theory although it should be noted that strictly this should only be used for thick films ( $t > \lambda$ ), whereas our data is better described by  $t \sim \lambda$ . This was also the case for Nagel et al [1994] who, nevertheless, were able to obtain good agreement between their data (from films of thickness comparable to the coherence length) and the theory of Werthamer.

Werthamer [1963] used the mathematical analogue of the energy levels of a particle in a onedimensional square potential well to obtain a set of coupled equations relating the transition temperature of the bilayer to fundamental parameters of the superconducting (s) and normal layers (n). These are: thickness of the layers,  $t_s$  and  $t_n$ , the effective coherence lengths of Cooper pairs,  $\xi_s$  and  $\xi_n$  (different range of electron interaction in the normal and superconducting metals), characteristic lengths,  $k_s$  and  $k_n$  (or depth of electron penetration) and critical temperatures  $T_{cs}$  and  $T_{cn}$  and  $T_c$ . The superconducting critical temperature,  $T_c$  is the transition temperature of the bilayer and  $T_{cs}$  is the temperature at which a pure iridium film becomes superconducting (for a sputtered film deposited at room temperature,  $T_{cs} = 289$  mK [Fukuda, D., 2000]). If the corresponding temperature for the normal silicide layer,  $T_{cn} = 0$ , then Werthamer's equations reduce to [Nagel, U., 1994]:

$$k_n = \frac{1}{\xi_n} \tag{7-14}$$

$$\ln\left(\frac{T_{cs}}{T_c}\right) = \aleph\left(k_s^2 \xi_s^2\right) \tag{7-15}$$

$$\ln\left(\frac{T_c}{T_{cn}}\right) = -\aleph\left(-k_n^2 \xi_n^2\right)$$
(7-16)

$$k_s \tan(k_s t_s) = k_n \tanh(k_n t_n)$$
(7-17)

Where:

$$\aleph(z) = \psi\left(\frac{1}{2} + \frac{1}{2}z\right) - \psi\left(\frac{1}{2}\right)$$
(7-18)

and  $\psi$  is the digamma function, the derivative of the natural logarithm of the gamma function (first introduced by Euler to generalise the factorial to non integer values [Sibagaki, W., 1952]). Two alternative derivations of these equations are given by Werthamer [1963].

Equation (7-18) may be approximated by [Hauser, J., 1964]:

$$\Re(z) \to \ln\left[1 + \left(\frac{\pi^2}{4}\right)z\right] \qquad \text{for } z \ge 0 \qquad (7-19)$$
$$\Re(z) \to \left(\frac{\pi^2}{4}\right)\ln(1+z) \qquad \text{for } z \le 0 \qquad (7-20)$$

Equations (7-14) - (7-20) may be combined to determine the depth of normal silicide layer required to produce the measured reduction in transition temperature.

If we assume a thin normal layer such that  $t_n \ll \xi_n$  then equation (7-17) reduces to:
$$t_n = \xi_s^2 k_s \tan(k_s t_s) \tag{7-21}$$

Where  $t_s$  is known and  $k_s$  is determined by substitution of equation (7-20) into equation (7-15):

$$k_{s} = \left(\frac{2}{\pi\xi_{s}}\right) \left[\left(\frac{T_{cs}}{T_{c}}\right) - 1\right]^{\frac{1}{2}}$$
(7-22)

Substituting equations (7-22) and (7-13) back into equation (7-21) yields an estimate for the depth of the normal layer:

For sample #5 ( $T_c = 52 \text{ mK}$ ),  $t_n = 3.2 \pm 0.2 \text{ nm}$ For sample #6 ( $T_c = 157 \text{ mK}$ ),  $t_n = 0.6 \pm 0.03 \text{ nm}$ 

These errors are dominated by uncertainties in measurement of  $T_c$  and the superconducting layer thickness. If the error in measurement of  $T_c$  is increased by a factor of two, the error on  $t_n$  increases by 39 %. Similarly, if the error in the measurement of the iridium layer thickness is doubled to 10 nm, the effect on the error of  $t_n$  is an increase of 75 %. This analysis reveals how the addition of a very thin silicide layer (~ 3% of the iridium film thickness) may reduce the transition temperature from 289 mK to 52 mK. The suppression in  $T_c$  as a function of the normal layer thickness is plotted in figure 7.10 and is best described by a linear fit. This is expected from the de Gennes – Werthamer theory in the regime of thin normal layers [Nagel., 1994].

Also plotted in figure 7.10 is an estimate for the extent of the silicide layer as a function of the deposition temperature. This (red) curve is based on a line of best fit obtained from a combination of our data and that of Fukuda et al. [2000] and is based on the assumption that the deposition rates (and therefore the time exposed to elevated temperatures) were identical for all samples. The Leicester data set used to estimate the relation between deposition temperature and transition temperature was reduced from that shown in figure 7.8 to only include samples of 100 nm thickness, and is reproduced in figure 7.10, inset. A quadratic curve fits the data well, but it should be noted that the data set is small and, pending further experimentation, any analysis based upon this fit should be regarded as tentative. The transition temperature suppression is linear with the depth of the normal layer (blue curve in figure 7.10). This suppression shows a quadratic dependence on deposition temperature (red curve in figure 7.10).



**Figure 7.10**: The suppression of transition temperature with increasing normal layer thickness (blue curve, left hand scale) and an estimate of the corresponding relation between normal layer thickness and deposition temperature (red curve, right hand scale). Inset – data from which a quadratic dependence of transition temperature on deposition temperature was ascertained. If a logarithmic fit is used, the growth of the silicide layer becomes exponential with deposition temperature.

#### 7.7 Conclusion

In summary, the transition data obtained for our iridium films is consistent with that of other groups and indicates that iridium typically has a higher transition temperature in thin films than in the bulk state. The transition may be tuned, primarily by variation in the deposition temperature. This may be interpreted as controlling the rate of growth (and hence, thickness) of a thin normal silicide layer which forms during deposition and acts on the superconductor via the proximity effect. The model of de Gennes – Werthamer has been applied to this system. This suggests silicide layer thickness of up to 3 % of the superconductor thickness is sufficient to produce the observed suppression in transition temperature. Alternative models exist to describe the proximity effect and to predict the normal layer thickness. However, we found Chen's model [1990] to be a poor fit to our Al/Ag data and the electron diffusion model of Usadel [1970] requires additional information on the density of states and coherence length in the normal layer and resistance at the interface between the two metals, which is not readily available for such ill-defined IrSi<sub>x</sub> layers.

# **Chapter 8**

# **Digital signal processing**

#### 8.1 Experiment Description

In June 1999, prior to the fabrication of a UK TES detector, we were invited to join our colleagues at the Space Research Organisation of the Netherlands (SRON, Utrecht) during X-ray testing of one of their TES detectors. They also operate an Oxford Instruments KelvinOx 100 liquid helium dilution refrigerator for characterisation of their TES X-ray microcalorimeters. At Leicester, we have developed a digital data acquisition system and this was used alongside SRON's analogue readout electronics during X-ray data acquisition. Thus, we were able to make a direct performance comparison between digital and analogue signal processing methods.

## 8.1.1 TES Detector

The SRON TES consisted of a Ti/Au thermometer with a 500 x 500 x 0.5  $\mu$ m Cu absorber and superconducting aluminium tracking, deposited on a 1  $\mu$ m thick Si<sub>3</sub>N<sub>4</sub> membrane (figure 8.1). The expected intrinsic energy resolution was approximately 11 eV FWHM at 6 keV, but this could not be realised during this particular experiment run where resolution was system noise limited. The previous SRON design iteration incorporated a thicker (2  $\mu$ m) Cu absorber and achieved 25 eV FWHM energy resolution at 6 keV. Subsequent designs have further improved on these prototypes (12.4 eV FWHM measured at 6 keV [Hoevers, H., 2000]). Readout in each case was via a Hypres SQUID (see Chapter 3) and input X-rays were generated by an <sup>55</sup>Fe source mounted in the refrigerator.

#### 8.2 Signal Processing

Pre-amplification and X-ray signal processing are vital components in an X-ray spectrometer. For laboratory development systems, analogue electronics are convenient and frequently used; a space instrument, however, will almost certainly require the potential performance gain (both in terms of energy resolution and spurious event discrimination) possible with digital processing [Knoll, G., 2000]. With a view to future detector applications and to maximise energy resolution in the presence of noise, we have developed a digital signal processing chain for use in the laboratory.



**Figure 8.1**: Optical micrograph of a development prototype SRON Ti/Au TES detector. The copper absorber overlays the aluminium bias rails and TES. The detector is deposited in the centre of a 3 x 3 mm square  $Si_3N_4$  membrane.

#### 8.2.1 Filter Theory (FFT)

The purpose of the signal processing electronics is to effectively filter noise from the detector signal and accurately determine the energy resolution of the detector. Fourier techniques are routinely employed to remove noise from corrupted signals [Gonzalez, R., 1987]. In this instance, the output pulses from the SQUID form the underlying signals u(t) to be measured. For any non-perfect detector, each signal pulse becomes modified by convolution with the detector impulse response function r(t)resulting in a smeared signal, s(t) [Proakis, J., 1996]:

$$s(t) = \int_{-\infty}^{\infty} r(t-\tau)u(\tau)d\tau$$
 or

$$S(f) = R(f)U(f)$$
(8-1)

Where the response function is assumed to be centred on t = 0. A delta function feature in the input signal  $u(\tau)$  occurring at time  $\tau$  is smeared into the shape of the response function and translated from time t = 0 to time  $\tau$ . S, R and U are the Fourier transforms of s, r and u, respectively.

Deconvolving the effects of the response function is a simple matter of dividing S(f) by R(f). However, in practice, the smeared signal s(t) is further degraded by the addition of noise, n(t) to form a measured corrupted signal, c(t) [McDonough, R., 1995]:

$$c(t) = s(t) + n(t) \tag{8-2}$$

In the presence of a noise component, it is necessary to apply a filter  $\phi(t)$  or  $\Phi(f)$  to determine the original signal. The filter is optimised when it yields a signal V(f) that is as close as possible to the original signal U(f), when applied to the measured signal C(f) and deconvolved by R(f),:

$$V(f) = \frac{C(f)\Phi(f)}{R(f)}$$
(8-3)

An optimum (Wiener) filter implies minimising the least-square difference between the original and estimated signals [Lyons, R., 1997]:

$$\int_{-\infty}^{\infty} \left| v(t) - u(t) \right|^2 dt = \int_{-\infty}^{\infty} \left| V(f) - U(f) \right|^2 df$$
(8-4)

Substituting equations (8-2) and (8-3) in (8-4) gives:

$$\int_{-\infty}^{\infty} \left| \frac{\Phi(f)[S(f) + N(f)]}{R(f)} - \frac{S(f)}{R(f)} \right|^2 df$$
(8-5)

Differentiating the integrand with respect to  $\Phi(f)$  and setting the result to zero minimises the least-squares difference and yields the formula for an optimum filter [Press, W., 1992]:

$$\Phi(f) = \frac{|S(f)|^2}{|S(f)|^2 + |N(f)|^2}$$
(8-6)

In order to know the correct form for the optimum filter, it is necessary to estimate the noise power spectrum  $|N(f)|^2$  and smeared signal spectrum  $|S(f)|^2$  separately. This is best achieved by taking a long sample measurement of the noise only and subtracting this from the measured corrupted signal power spectrum  $|C(f)|^2$  in order to estimate the smeared signal spectrum  $|S(f)|^2$ . Fortunately, since the optimum filter results from a minimisation problem, the quality of the results obtained differ from the true optimum by an amount that is second order to the precision to which the optimum filter was determined. An advantageous consequence of which is that the measured energy resolution of the detector does not depend critically upon the rigorous determination of the optimum filter shape.

# 8.2.2 Line Broadening Mechanisms

The discussion above assumes that all of the spectral line broadening results from signal corruption due to instrumentation and noise effects. This is not the case. A number of physical processes may act to account for the non-zero width of an astronomical spectral line, the most important include natural broadening, Doppler broadening and collisional broadening [Griem, H., 1974].

Natural line broadening occurs since neither atomic levels nor the lines connecting them are infinitely sharp [Hollas, J., 1996]. This may be explained by the uncertainty principle:  $\Delta E \Delta t \sim \hbar$ . An atom in a state for duration  $\Delta t$ , implies a finite spread in energy,  $\Delta E$ . This results in the Lorentz line profile [Hollas, J., 1996]. Lorentz profiles may also be generated if the atom suffers collisions at random frequency with other particles while it is emitting.

Doppler broadening is an important feature of astronomical observations (Chapter 1) and arises from the thermal motion of the emitting (or absorbing) atom. The Doppler shift is of random direction, so the net effect is to spread the line out in a Gaussian distribution but not to change its total strength.

In the laboratory context where X-ray sources are sealed (typically <sup>55</sup>Fe samples), only natural line broadening applies. The magnitude of natural line broadening for stationary sources is typically small  $(2.13 \pm 0.19 \text{ eV} \text{ for the Mn-K}\alpha$  line [Lee, P., 1974]) and is often neglected in spectral analysis, but becomes significant for cryogenic detectors capable of intrinsic energy resolutions < 10 eV. For this experiment, natural broadening was only a small fraction of the measured instrument energy resolution (section 8.3), and was not accounted for in the current data analysis. This should be addressed as the energy resolution of the detector is improved.

#### 8.2.3 Data Acquisition Hardware

The hardware used to acquire X-ray pulses consisted of a 16-bit ADCDSP (Analogue-to-Digital Converter Digital Signal Processor) developed jointly by Xcam Ltd (20 York Rd, Northampton, UK) and Leicester University. Output signals from the SQUID were input to an Analog Devices (One Technology Way, P. O. Box 9106, Norwood, MA 02062-9106, USA) AD1671 1.2 MHz 12-bit ADC. A 4096 word FIFO (First In First Out) buffer resides between the ADC and an Analog Devices ADSP2181 DSP. Digitised data were retrieved by the PC via the ISA (Industry Standard Architecture) bus.

The analogue input signal could be triggered either continuously, in order to record noise from the system, or triggered by signals rising above a threshold value. The latter method was used to identify the rising edge of an X-ray pulse and initiate data transfer to the DSP.

# 8.2.4 Data Acquisition Software

SRON shaped the detector output pulses using an analogue integrating RC-filter. In the absence of noise, or in the presence of white noise, this produces similar results to a digital approach since Fourier analysis offers little gain in resolution under these conditions. The ability to fit multiple Gaussian profiles to a spectrum and measure the confidence in the fit (compared to fits using a single Gaussian) via an F-test [Patel, J., 1996] is an important feature of the software developed at Leicester, although it remains untested (all data sets to date have been well described by single Gaussians).

The graphical user interface of the software package was written in Visual Basic, with computational algorithms written in C++. The software is able to record a sample of raw data and plot the Fast Fourier Transform (FFT) of noise. Individual pulses may be plotted or saved to a file. The optimum filter is derived from the noise power spectrum and convolved with a given data file and the results plotted as a histogram.

Energy resolution of a Gaussian peak is obtained by fitting the data by method of maximum likelihood (a general method of which least-squares fitting is a simple formulation) [Fisher, R., 1922]. The principle behind maximum likelihood is that given an estimate for the source function, it is possible to estimate the probability (or probability density) of observing a particular event in the data set. In general, a likelihood function, L is constructed from the product of the individual probabilities. The maximum likelihood occurs when L is stationary with respect to changes in the estimate function or the fitting parameters. This is straightforward for a linear data set [Wolberg, J., 1967], but since a Gaussian distribution is non-linear, an explicit best-fit solution to the Mn-K $\alpha$  peak, for example, cannot be found. Therefore, in order to provide the best estimate for pulse amplitude, the software uses a Marquardt approach [Marquardt, D., 1963] which combines a grid search for initial estimates of the Chi-square value with a subsequent gradient search on the generated Chi-square profile to locate global minima. Fitting of a parabola to the Chi-square curve accurately determines the best estimate for the mean value of the parameter. By minimisation of the Chi-square value, a best fit is achieved for the Gaussian profile.

The software includes the capacity to define a region of interest in the spectrum, apply the fitting algorithm and calculate a Gaussian fit to the chosen data. By specifying a limited data set, anomalous tails on Gaussian profiles may be discounted from the computation of the underlying FWHM value. This is a simple method of event discrimination. In future, such selection procedures based on pulse profile could be automatically programmed into the software.

#### 8.3 Results and Discussion

In real-time during data acquisition at SRON, the software was used to plot the FFT of a sample of system noise. This showed a flat frequency response, permitting the use of a relatively simple filter: an average of all pulse shapes. Inspection of equation (8-6) shows that the filter tends to unity in the absence of noise, and zero if noise is dominant. For an intermediate case, if 1/f noise was present for example, the optimum filter deviates from the average pulse shape and takes the form shown in figure 8.2.



**Figure 8.2**: Left, a suitable filter shape in the presence of 1/f noise. If the noise is white, this filter may be replaced with an average pulse profile, such as that shown on the right.

For the duration of the experiment, a frequent shift in the baseline of the SQUID flux-locked-loop (FLL) was observed, apparently due to an occasional X-ray event causing the SQUID to lock on to a different part of the V- $\Phi$  curve. This continued until the baseline shifted beyond the range of the analogue-digital converter (ADC). To counter this, an automatic reset on the ADC was implemented using an optic coupler. The reset time was set to 150 ms (some double resets were observed when set to 100 ms) with a 500 ms delay before acquisition of data. This successfully corrected the drift for the ADC, but instabilities in the SQUID FLL remained a limiting factor on longer duration acquisition of spectra with improved counting statistics: each spectrum was limited to less than one hour duration.

Despite SQUID instabilities, a number of spectra were acquired; an example is shown in figure 8.3. The raw data has been convolved with a filter constructed from the average pulse shape and plotted using the data processing software. However, the spectra obtained were sufficient to confirm the FWHM energy resolution of the detector as measured by SRON and characterise the performance of the digital data acquisition electronics. As expected in the presence of white noise, the digital system did not significantly reduce the energy resolution estimate for the detector but was invaluable when investigating spectral anomalies.



**Figure 8.3**: Spectrum from <sup>55</sup>Fe source using a Ti/Au SRON detector with a digital pulse processing system developed at Leicester University. Gaussian fitting to the Mn-K $\alpha$  peak gives an energy resolution FWHM of 25 eV. The additional peak (red) to the right of the main Mn-K $\alpha$  peak arises from suspected photon interactions in the TES itself.

In the spectrum shown in figure 8.3, the Mn-K $\alpha$  and K $\beta$  peaks are unambiguously resolved. Calibration of the energy scale is taken from the relative position of these peaks. The FWHM value of 25 eV for the Mn-K $\alpha$  peak was typical of spectra obtained during the experiment, and very similar to results from spectra acquired using analogue signal processing. Interesting features in the spectrum include the broad spread of counts between 5300 eV – 5800 eV (possibly substrate events) and also a smaller anomaly at higher energies (shown in red in figure 8.3). To investigate the source of these spectral features, the shapes of pulses from each region were compared using our software. Pulses from all energies up to, and including, the Mn-K $\alpha$  peak showed a high degree of uniformity and an average pulse shape is shown in yellow in figure 8.4. In contrast, pulses from the high energy anomaly (~ 6000 eV) showed distinct baseline and rise time differences (red in figure 8.4).

Faster risetimes and slightly larger pulse amplitudes are indicative of events occurring in the TES rather than the absorber. When an X-ray is absorbed directly in the TES, a primary photoelectron is generated which causes a rapid electron collision cascade [Ukibe, M., 2000]. Heating of the electron system increases the TES resistance. This thermalisation process is faster than the equivalent process in the absorber, hence the faster risetime. Close thermal coupling between the absorber and TES ensures efficient heat transfer from the TES to the absorber and so the decay times are similar for absorption in the TES or absorber. Such effects can be removed either by increasing the spatial restriction of the X-ray collimator or by post-processing event rejection.



**Figure 8.4**: Comparison of pulse profiles between events from the Mn-K $\alpha$  photopeak and events from the anomalous region centred around 6000 eV, highlighted in figure 8.3. The principal difference is in risetime, and a slight baseline shift is also evident. Faster risetime is characteristic of photon absorption in the TES.

## **8.4 Conclusion**

We have demonstrated the use of a digital data acquisition system for processing X-ray pulses and reproduced energy resolution results obtained from an analogue chain. The advantage of a digital approach over conventional analogue electronics became apparent when investigating anomalous spectral features. We were able to begin to determine a range of instrumental artefacts such as pulse risetime effects and detection arising from sources other than the absorber, and apply spurious signal rejection.

The software is still under development and will be used for future X-ray testing of Oxford Instruments TES detectors in Leicester. It is expected that this will not only accurately record X-ray spectra but also provide a more powerful insight into the physics of the operation of these devices than is possible with an analogue process chain.

# **Chapter 9**

# Large absorber imaging detectors

# 9.1 Alternative Designs for Imaging Detectors

Currently, two types of design are being considered for realising an imaging TES detector. Most groups (for example SRON [Bruijn, M., 2000a] and the National Institute of Standards and Technology (NIST), Colorado [Deiker, S., 2000]) are pursuing arrays of independent TES pixels. If the technical challenges can be overcome, this is likely to prove the best solution for many astronomy applications. Such a pixellated detector would have spatial resolution limited only by the dimensions of the absorber pixels (and associated packing efficiency), energy resolution comparable with single pixel devices and high countrate capability since incident events are distributed across many pixels. However, substantial investment is required to resolve the outstanding technical difficulties with this scheme, such as multiplexing SQUID readout to simultaneously read all pixels and eliminating thermal cross-talk between pixels. Presently, proposed detector configurations are generally complicated and require significant improvements in fabrication techniques [Bruijn, M., 2000b].

A simpler alternative is to employ a single, large absorber with multiple TES readout (dubbed by researchers at ESTEC in the context of STJ sensors the Distributed Read-Out Imaging Device, or DROID concept [den Hartog, R., 2000]). An alternative US acronym is PoST: Position-Sensing Transition-edge sensor [Figueroa-Feliciano, E., 2000]. The relative timing and amplitude of heat pulses in each TES yields position information about the X-ray event. Several absorbers may be tessellated to fill the focal plane. Such a common absorber configuration has a lower maximum countrate than a pixel array but benefits from much simpler signal readout (two channels per axis). As a consequence, there is the potential for more sophisticated digital filtering and pulse processing, important for spacecraft with restricted telemetry resources. One disadvantage of this layout is that a large absorber must have a correspondingly large heat capacity, degrading energy resolution. Dielectric and superconducting absorbers are a possible solution but are not favoured for reasons outlined in Chapter 2. Our design proposals utilise bismuth absorbers. The heat capacity of bismuth at 100 mK is ~ 40 times lower than that of gold, facilitating in principle the use of larger absorbers without compromising energy resolution. The low heat capacity is due to the lack of free electrons in bismuth, another (unwelcome) consequence of which is that bismuth has relatively poor thermal conductivity (~ 86 Wcm<sup>-1</sup>K<sup>-1</sup> at 3.5 K [Issi, J-P., 1977], dominated by the phonon contribution). For a large area absorber, it is important that heat transfer to the TESs is fast, otherwise the countrate (already compromised by the use of a large common absorber) may be too low for a useful imaging

detector. Hence, understanding the mechanisms of heat flow in bismuth, and phonon propagation in particular, is essential before a detector can be designed.



# 9.2 An Experiment to Investigate Heat Transfer in Pure Bismuth

**Figure 9.1:** Proposed mask design for testing the thermal conduction properties of bismuth at low temperatures. The design utilises a standard square SRON membrane and incorporates the fabrication tolerances of the SRON facility (error margins for overlaying mask sets, for example).

An experiment is proposed to test the principle of a large area bismuth absorber, with fabrication and testing scheduled for late 2001. Figure 9.1 shows the layout of our test device. It consists of a linear strip of bismuth, 300  $\mu$ m wide, 20  $\mu$ m thick and 4000  $\mu$ m long arranged to fit inside the maximum dimensions of a Si<sub>3</sub>Ni<sub>4</sub> membrane fabricated by SRON (3 mm x 3 mm). A Ti/Au TES is coupled to each end of the bismuth absorber. Heat pulses to simulate X-ray absorption events are generated by applying voltage pulses to copper heaters at various positions along the bismuth strip. Contact to these heaters is made with aluminium tracks leading to wire-bond pads. Three alignment marks are shown, designed to ensure correct alignment of each of the six masks.

The time delay between pulse injection and measurement by the TESs and consideration of the measured pulse shapes (and how these vary with distance propagated through the bismuth) may be compared to results from a model. It is expected that this experiment will improve our understanding

of heat transport mechanisms in bismuth and verify predicted trade-off relations between countrate and maximum length of bismuth absorber. An estimate may also be made of the spatial resolution limit imposed by the limit of discrimination of the difference in pulse amplitudes and time of arrival of pulses at each TES. In a subsequent experiment, we intend to investigate how transport of heat from an X-ray event to the TES could be improved by the addition of a thin copper layer to the bismuth surface. It is possible to grow thin metal films that are acoustically well coupled to bismuth [Kashirin, V., 1994] and it is expected that this will increase the speed of response without significantly increasing the heat capacity of the absorber.

Unfortunately, problems with our dilution refrigerator have resulted in delays to our experimental programme and these experiments have not been conducted in time for submission of this thesis. However, heat transport in bismuth has been extensively modelled; the results are discussed in section 9.5.

## 9.3 Physics of Heat Flow in Bismuth at Low Temperatures

#### 9.3.1 Properties of Bismuth

Bismuth is a compensated semi-metal with a carrier density of ~ 3 x  $10^{17}$  cm<sup>-3</sup>, as opposed to ~ $10^{22}$  cm<sup>-3</sup> for a normal metal and ~ $10^{11}$  cm<sup>-3</sup> for silicon [Cassignol, E., 1966]. This is equivalent to a free carrier density of  $10^{-5}$  per atom [Pratt, W., 1978]. It consists of a single stable isotope and so there is no isotopic scattering of phonons which limits the thermal conductivity of many other solids at low temperatures [Narayanamurti, V., 1972]. Phonons are also scattered by other phonons or by charge carriers. Microscopic stacking faults in the crystal lattice or impurity atoms also constitute scattering centres. At low temperatures, scattering of phonons by the surface of the crystal is significant. The dominant type of scattering (and hence the mechanism determining the thermal conductivity for T < 20 K) is strongly temperature dependent.

#### 9.3.2 Thermal Conductivity of Bismuth as a Function of Temperature, $\kappa(T)$

In bismuth, the electron and phonon contributions to thermal conductivity are approximately equal for  $T \ge 120$  K [Kagan, V., 1991]. As the temperature is reduced, the electron component reduces linearly with T and is insignificant for T < 20 K [Mezhov-Deglin, L., 1975]. Thermal conductivity (phonon mean free path) is then determined by scattering processes of the phonons. In a pure bulk bismuth sample the phonon thermal conductivity reaches a maximum at a temperature of 3.5 K [Issi, J-P., 1977]. At this temperature, phonon-phonon scattering dominates and the motion of phonons is described by a Poiseuille flow [Mezhov-Deglin, L., 1975]. This resembles a 'random walk' Brownian

motion of a particle or the flow of viscous liquid in a pipe under the influence of an external force. Below this temperature, scattering of phonons by electrons increases (and hence thermal conductivity,  $\kappa$  decreases) with decreasing temperature.

$$\kappa \propto T^3 \qquad \text{for } T < 3.5 \text{ K.} \qquad (9-1)$$

Electron scattering occurs only for phonons with thermal momentum less than or equal to the transverse dimension in **k** space of the electron (or hole) ellipsoid, 2  $p_F$  [Red'ko, N., 1994]. In bismuth,  $p_F \sim 5 \ge 10^{-22}$  gcms<sup>-1</sup> and 15  $\ge 10^{-22}$  gcms<sup>-1</sup> for electrons and holes, respectively [Mezhov-Deglin, L., 1975]. At around 0.3 – 0.5 K the characteristic momentum of the thermal phonons becomes comparable with 2  $p_F$  and a corresponding minimum in effective phonon mean free path  $l_{eff}$  has been observed [Mezhov-Deglin, L., 1975] – an effect first described by Ziman [1960].

For T < 0.3 K, the phonon mean free path increases and at lower temperatures the only significant scattering of phonons is by the surfaces of the sample, leading to ballistic (Knudsen) flow. This is the regime in which our detectors will operate ( $T \sim 100$  mK).

#### 9.3.3 Knudsen Flow ( $T \leq 0.1$ K)

Knudsen flow is defined by the condition [Red'Ko, N., 1991]:

$$d < l_N < l_U \tag{9-2}$$

Where d is the minimum sample dimension and  $l_N$  and  $l_U$  are the mean free paths of normal (N) and Umclapp (U) phonons, respectively.

In any solid, the only reason for phonon-phonon interaction is the anharmonicity of the lattice vibrations (inelastic deformation of the lattice) which results in collisions of two types: normal (N) and Umklapp (U) processes, each with characteristic time constants. For N processes, the total energy and momentum of the phonon system is conserved whereas U processes include dissipation of energy  $(\hbar\omega)$  and momentum  $(\hbar k)$  and contribute to thermal resistivity.

For N processes:

Energy:  $\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$  (9-3)

Momentum:  $\hbar \underline{k}_1 + \hbar \underline{k}_2 = \hbar \underline{k}_3$  (9-4)

Whereas, for U processes:

$$\hbar \underline{k}_1 + \hbar \underline{k}_2 = \underline{\hbar} \underline{k}_3 + \hbar \underline{G} \tag{9-5}$$

where G is a reciprocal lattice vector.

By this definition, U processes include scattering of phonons by carriers, crystal stacking faults and sample surfaces. The temperature dependence of the average mean free paths for N and U processes in bismuth has been determined experimentally [Kopylov, V., 1974]:

$$l_U = s\tau_U = 5 \times 10^{-5} e^{\left(\frac{40}{T}\right)} \text{ cm}$$
(9-6)  
$$l_N = s\tau_N = AT^{-4} \text{ cm}$$
(9-7)

where A = 0.4 to 3 [Kuznetsov, M., 1970 and Narayanamurti, V., 1972, respectively] and the average sound velocity,  $s = 1.5 \times 10^5$  cms<sup>-1</sup> [Kagan, V., 1991].

In general, the temperature dependence of the mean free paths  $l_N$  and  $l_U$  dictates the type of phonon trajectory at a given temperature. At very low temperatures and reasonable sample dimensions (e.g. T < 0.1 K and  $d = 100 \,\mu$ m) the condition of (9-2) is met. This is the Knudsen flow regime where phonons driven by a temperature gradient move between boundaries of the sample, rarely colliding in the bulk of the crystal. The effective mean free path of such phonons then increases slightly with decreasing temperature, tending asymptotically to d. In general, Knudsen flow is characterised by a time constant of  $\tau_d \sim d/s$ .

In this regime the thermal conductivity,  $\kappa$  is described by [Red'Ko, N., 1991]:

$$\kappa \approx \frac{1}{3} Csl_{eff} \approx Ad \left(\frac{T}{\theta}\right)^3$$
(9-8)

where C is the phonon specific heat in the Debye approximation (~ 5.5 x  $10^{-5} T^3$  Jcm<sup>-1</sup>K<sup>-4</sup>) and  $\theta$  is the Debye temperature of bismuth (120 K [Kagan, V., 1993]).

The thermal conductivity is governed entirely by the scattering of phonons by boundaries; known as the Casimir thermal conductivity mechanism [Red'Ko, N., 1994]. Assuming diffuse scattering at boundaries, the corresponding effective Casimir thermal conductivity can be deduced [Kagan, V., 1992].

In summary, since a TES is expected to operate at approximately 100 mK, heat transport in a bismuth absorber is by ballistic Knudsen flow, with some scattering of phonons by electrons possible. All

other phonon scattering processes (with the exception of surface scattering) may be neglected in any analysis.

## 9.4 Modelling

# 9.4.1 Alternative Modelling Approaches

In this Chapter, a Monte Carlo simulation of individual phonon trajectories in a low temperature bismuth absorber is described. This approach differs from previous models of cryogenic detectors which have concentrated on solving the diffusion equation [Hanna, A, 1996] or the generalised heat conduction equation [Deptuck, D., 1996]. The model of Deptuck et al. [1996] generated model pulses which were qualitatively similar to observed pulses, but these authors were unable to obtain a good fit for both pulse height and shape simultaneously. They conclude that although diffusion effects are important, such processes do not fully account for device behaviour. Hanna et al. [1996] apply the diffusion equation (in two dimensions only since the detector is fabricated on a membrane) to a 100 x  $100 \ \mu\text{m}^2$  gold absorber with a single normal-insulator-superconductor readout. They show how errors may be caused if photon energy is determined by pulse height only. A numerical correction based on either the rise time or fall time of the pulse was used to deduce both the photon energy and position in the absorber. Results from our Monte Carlo model (figure 9.3, for example) are in good agreement with those of Hanna et al. [1996]. Our approach also yields a greater insight into the fundamental physics at a microscopic level.

## 9.4.2 The Monte Carlo model

A Monte Carlo simulation of heat flow in a low temperature bismuth absorber has been developed to investigate spatial resolution in extended bismuth absorbers. The programme was written in C++. Code by Matsumoto [1998] for pseudorandom number generation was used to minimise the probability of patterns in the 'random' numbers corrupting the results, and to improve the speed of execution.

The ballistic trajectories of up to 30000 phonons (limited by computer memory constraints, but sufficient to yield smooth TES responses) originating from a specified point of X-ray absorption were tracked through a 3-dimensional bismuth absorber from the point of thermal excitation to readout at a TES. The phonons were assigned a random initial direction and velocity of 1500 ms<sup>-1</sup>, the mean sound velocity in pure bismuth [Kagan, V., 1991]. Scattering at absorber boundaries was elastic with an option to include absorption of the phonon at the boundary. Each phonon was tracked until absorbed in a TES. The output pulse from the TES was generated with the assumption that the instantaneous

TES temperature is proportional to the number of phonons entering the TES. Inherent in this assumption are the conditions that the phonon spectrum produced by the X-ray absorption is completely monoenergetic and that there were no upscattering or downscattering processes which can change the phonon energy. Furthermore, it becomes necessary to assign phonons an initial velocity only, not an energy. With the exception of surface absorption effects, no heat sinks have been included in the model. Therefore, the energy of the incident X-ray may be deduced from the sum of the integrals of the TES pulses and it is possible to deconvolve the energy and position information (assuming a suitable energy calibration). A phonon absorption probability of unity at the bismuth/TES boundary is unrealistic. Following the work of Irwin [1995b], a value of 0.3 has been assumed, although the results do not depend critically upon this parameter. Irwin [1995b] deduced this value for phonon absorption at a silicon absorber/tungsten TES boundary by fitting modelled phonon propagation results to experimental data.

A baseline temperature value of T = 100 mK has been assumed. This places the phonon system in a state of Knudsen flow where boundary scattering is the dominant contribution to thermal resistance and electron scattering is minimal. Phonon reflection at boundaries is taken to be diffusive. This fits observed data well [Kopylov, V., 1974] and is equivalent to treating the phonons as black-body radiation which is absorbed at the boundary and re-emitted as if from a perfectly rough surface with no directional dependence. In the programme, the emission angles are chosen from a uniform random number distribution. As shown by Casimir [1938], the effective mean free path of such phonons then increases slightly with decreasing temperature, tending asymptotically to 1.12d, for a square section of side d [Ziman, J., 1960]. The proportion of phonons that are specularly reflected may increase as the temperature decreases. This effect arises from the apparent improvement in the surface polish as the phonon wavelength increases. However, this effect is considered negligible when considered in context with electron scattering [Mezhov-Deglin, L., 1975], and so has not been included in the model.

#### 9.5 Model Results

### 9.5.1 Linear Bismuth Absorber

Various detector geometries have been modelled. The results presented in this section are for a linear bismuth strip (length (L) 4000  $\mu$ m x 300  $\mu$ m (width) x 22  $\mu$ m (thickness)) with a TES at each end (figure 9.2). This simulates the results from the experiment outlined in section 9.2, and demonstrates the linear spatial resolution that may be obtained by comparison of pulse amplitudes in each TES.



Figure 9.2: Schematic linear bismuth absorber geometry. Note the two TESs at x = 0 and x = L and the variable X-ray absorption position. Not to scale.

A bismuth absorber thickness of 22  $\mu$ m was chosen as an upper limit since this corresponds to a quantum efficiency of 95 % at 10 keV (Chapter 2). This size of absorber has a specific heat capacity of 4.03 x 10<sup>-12</sup> J/K, the same order of magnitude as for the gold absorber in the numerical example of Chapter 2 (which resulted in a thermodynamic energy resolution limit of 2.5 eV). To optimise energy resolution, thinner films could be used without significantly compromising quantum efficiency, but in the Knudsen phonon flow regime this gives rise to more internal boundary scatter before phonon absorption in the TES. Given the diffuse nature of boundary scattering in bismuth at these temperatures, this leads to longer pulses and reduced countrate capability.

Figure 9.3 is a plot of pulses generated in each TES for an X-ray absorption at  $x = 1200 \ \mu m$  (x/L = 0.3) and centralised in y and z dimensions. The deviations from smooth curves arise due to statistical fluctuations in phonon counting. This plot clearly shows that pulse shape depends upon distance from the TES. This may complicate the energy analysis since an average pulse shape filter may not be applicable for long absorbers.





To test the validity of this Monte Carlo simulation, the calculated rise times (time to reach maximum temperature,  $T_{max}$ ) may be compared to the analytical theory of Hanna, et al [1996], who state that the rise time of the pulse,  $t_r$  in a gold absorber is given by:

$$t_r = \frac{x^2}{4D} \tag{9-9}$$

where D is the thermal diffusivity,  $D = \frac{\kappa}{C}$ .  $\kappa$  is the absorber thermal conductivity and C is the specific heat per unit volume of the absorber, both assumed to be independent of temperature.

Therefore, the analytically predicted ratio of rise times is:

$$\frac{t_r \left(\frac{x}{L} = 0.3\right)}{t_r \left(\frac{x}{L} = 0.7\right)} = \frac{0.3^2}{0.7^2} \approx \frac{1}{5}$$
(9-10)

From figure 9.3, if the rise times are taken to be 7  $\mu$ s and 28  $\mu$ s respectively, then the equivalent ratio for the modelled rise times is:

$$\frac{t_r \left(\frac{x}{L} = 0.3\right)}{t_r \left(\frac{x}{L} = 0.7\right)} = \frac{7}{28} = \frac{1}{4}$$
(9-11)

This is in close agreement with the analytical result of equation (9-10), thereby providing a measure of confidence in our model.

However, inspection of similar curves for other X-ray absorption positions indicates that most of the spatial information is contained within the ratio of amplitudes, rather than in relative pulse timing. This may not be the case for thinner films with more surface scattering and correspondingly longer pulse rise times, which further compromise countrate. Therefore, to estimate the spatial resolution of this configuration, the peak pulse height in TES 1 (x = 0) divided by the sum of the pulse heights in both thermometers is plotted against X-ray absorption position (0 < x < L) in figure 9.4. The gradient of this blue curve shows that the central region is of high sensitivity and the ends have poorer spatial resolution. An analogy may be drawn with charge division in resistive sheets as used in MCP (Micro Channel Plate) resistive anode readout [Fraser and Mathieson, 1981]. The blue curve of figure 9.4 is similar to the equivalent plot given by Fraser for resistive anodes, a further validation of this model. To put the blue curve in a practical context, the change in pulse amplitude in TES 1 over a distance of

250  $\mu$ m has been plotted (red curve, right axis). In other words, if system noise limits the sensitivity of relative pulse heights to, say, 5 %, then approximately 75 % of the absorber can be said to achieve spatial resolutions better than 250  $\mu$ m, which is the current specification for XEUS (Chapter 1). It is expected that the discrimination of pulse heights will exceed 1 % (corresponding to ~ 60 eV at 6 keV), therefore all but the extreme end regions of the absorber (which will be shielded to eliminate photon absorption events in the TES) should surpass the XEUS spatial resolution requirement.



**Figure 9.4**: Modelled data for pulse height in TES 1 normalised by the total measured signal summed from both TESs (blue curve, left hand axis). The red curve (right hand axis) is derived from the gradient of the blue curve and represents the difference in pulse amplitude in TES 1 for a photon absorption displacement of  $250 \,\mu m$ .

At the centre of the absorber (0.4 < x/L < 0.6), the data points represent X-ray absorption positions 200 µm apart (i.e.  $\Delta x/L = 0.05$ ). This is adequate spatial resolution for many astronomy applications, including XEUS. Therefore, for this detector geometry, if differences in the ratio of pulse amplitudes of ~1 % can be measured, then it is possible to obtain at least 200 µm resolution in the x axis. This equates to ~ 20 resolution elements in 1-d. The resolution in the y axis is governed by the width of the absorber, 300 µm in this instance.

Following the work of Irwin [1995b], a degree of surface phonon absorption was introduced into the model. Irwin modelled diffusive phonon propagation in large (500  $\mu$ m) silicon absorbers and noted that the fit to his data was improved by the addition to the model of a 15 % probability of phonon absorption at surfaces. This was attributed to a layer of adsorbed helium on the silicon originating from the inner vacuum chamber (IVC) of the dilution refrigerator. Adsorbed helium is an efficient

absorber of phonons [Irwin, K., 1995b] and may potentially reduce the TES signal to almost zero. Our absorber is much thinner than that of Irwin and hence has more scattering events associated with each phonon. Figure 9.5 shows the effect of the addition of a 1% probability of phonon absorption upon reflection at any of the exposed surfaces. Again, it may be shown that the ratio of rise times scales approximately as the square of the distance from TES to the point of X-ray absorption, as in (9-9).





From this plot it is clear that a 15 % probability of surface absorption cannot be tolerated in a thin film absorber. Even 1 % has a serious effect. It should be also be noted that adsorbed helium layers tend to grow with time. If precautions are not taken, as the liquid helium level in the dilution refrigerator drops during an experiment the upper surfaces of the IVC warm above 4 K, releasing helium gas into the IVC. The detector will be relatively cold and so attract much of this gas. This would have a large impact on the energy resolution since the integral under the TES output curves for a given energy X-ray event may change during the acquisition of a spectrum. Further investigation into the probability of phonon absorption by a surface layer of helium is therefore warranted.

The model can also be used to simulate the effect of adding a copper layer to the bismuth. As described in section 9.2, it is expected that a thin copper layer in good acoustic contact with the bismuth will reduce the response time of the detector. Phonons absorbed at the bismuth-copper interface excite the electron system in the copper and facilitate faster readout via diffusion of these electrons to the TES at each end of the composite absorber (if each TES is in good electrical contact with the copper layer). Thermalisation of this energy within the copper electronic system is typically fast compared to the phonon propagation time in bismuth [Irwin, K., 1998], giving rise to the improvement in signal rise time. Hence, spatial resolution is now determined primarily by the

characteristics of electron diffusion in thin film, low temperature copper. Our current experimental programme does not include plans to implement such a copper layer and so the model has been adapted to simply demonstrate the gain in signal and response time, not to yield a full analysis of the electron diffusion in copper.

In the model, a layer of copper was added to the upper surface of the bismuth absorber. As before, 30000 phonons were created by an X-ray absorption event in the bismuth layer. These propagate through the bismuth and eventually impinge on the copper-bismuth interface. The probability of phonon absorption by the copper was a free parameter (1 % in figure 9.6). If absorbed, the phonon trajectory was terminated and the electron temperature in the copper incremented. A pulse in electron temperature is therefore constructed from the rate at which phonons are absorbed by the copper. In practice, the falling limb of the electron temperature curve would be determined by the electron – phonon coupling coefficient in copper and losses to any cold heat sinks. In an actual detector, this pulse would be read out by the TES thermometers in addition to their phonon response, giving rise to two characteristic fall times for the pulse: a fast initial decay (due to the electron component) followed by a slower phonon decay. For the purposes of illustrating the effect on speed of response of including a copper layer, the electron temperature pulse is de-coupled from the response of the TESs to phonon absorption, which are plotted separately in figure 9.6.



**Figure 9.6**: Simulated pulses for an X-ray absorbed at  $x = 1600 \ \mu m$ ,  $y = 150 \ \mu m$ ,  $z = 11 \ \mu m$ , in a composite bismuth/copper absorber. The phonon response of each TES is shown together with a normalised curve for the increment in copper electron temperature. The actual amplitude of the copper pulse is an order of magnitude greater than that for TES 1.

The curves of figure 9.6 were computed assuming an absorption probability of phonons in the copper of 1%. Despite such a low probability, the proportion of the signal in the copper layer is an order of

magnitude larger than either TES, and moreover, peaks over significantly shorter timescales. This simulation suggests that the addition of a normal metal layer with high thermal conductivity (e.g. copper or gold) would significantly increase the speed of response of a linear detector fabricated primarily of bismuth, and consequently would increase the potential count rate of the detector.

## 9.5.2 Square Bismuth Absorber

One aim of our programme is to develop a 2-d bismuth absorber with multiple TES readout, or Distributed Read-Out Imaging Device (DROID) [den Hartog, R., 2000]. To simulate such a device, the model was adapted to the geometry shown in figure 9.7. Each of the four TESs extends 500  $\mu$ m (10 % of absorber of side 5000  $\mu$ m) along the y axis.



Figure 9.7: Geometry of the DROID model (not to scale). The bismuth absorber (with no copper layer) is read out by 4 corner TES thermometers. Each TES extends 500  $\mu$ m along the y axis, forming a collecting area of 500  $\mu$ m x 22  $\mu$ m.

The X-ray absorption position was varied over the surface of the bismuth layer and the response of each TES was recorded. Figure 9.8 shows examples of the pulses generated. The dominance of TES 2 in this instance occurs because the relative amplitude of a TES pulse has a highly non-linear relation with the distance to the point of absorption. This is illustrated in figure 9.9 which shows the signal in TES 2 as a proportion of the total phonon count (valid since a monoenergetic phonon spectrum was assumed, see section 9.4.2) plotted as a function of the position of the incident X-ray.



**Figure 9.8**: Pulses from corner TESs for an X-ray absorption position of  $x = 1000 \ \mu m$ ,  $y = 4000 \ \mu m$ ,  $z = 11 \ \mu m$ . The system 'thermalises' when all TESs record the same temperature after approximately 50  $\mu$ s, the same as for the linear absorber.



**Figure 9.9**: Percentage of the total measured phonon count at each X-ray absorption position recorded by TES 2. The proportion tends to 100 % as  $x \to 0 \ \mu m$  and  $y \to 5000 \ \mu m$ , as required.

As with the linear absorber, most of the spatial information is contained in the difference in pulse amplitudes rather than in the pulse time of arrival (figure 9.8). For this reason, the position in the x direction has been determined by the ratio of the sum of peak amplitudes of the TESs at  $x = 0 \mu m$ , and the total measured signal, with no account taken of time of arrival:

$$\gamma_{x} = \frac{\left(TES1_{amp} + TES2_{amp}\right)}{\left(TES1_{amp} + TES2_{amp} + TES3_{amp} + TES4_{amp}\right)}$$
(9-12)

Similarly, the position in the y direction is deduced from the ratio of the sum of amplitudes of the TESs at y = 0, and the total measured signal:

$$\gamma_{y} = \frac{\left(TES1_{amp} + TES3_{amp}\right)}{\left(TES1_{amp} + TES2_{amp} + TES3_{amp} + TES4_{amp}\right)}$$
(9-13)

The size of the change in these ratios between adjacent X-ray absorption positions may be interpreted as a limit on the spatial resolution of the detector. The amplitude ratios,  $\gamma_x$  and  $\gamma_y$  are plotted as a function of absorption position in figure 9.10(a, b).



Figure 9.10(a): Amplitude ratio,  $\gamma_x$  as a function of position. Note the similarity between the profile of the curve at y = 2500 µm and the blue curve for the 1-d case in figure 9.4.



Figure 9.10(b): Amplitude ratio,  $\gamma_y$  as a function of position.

The change in gradient of the function  $\chi$  gives rise to variation in position sensitivity over the surface. For example, consider the difference between the gradient of the line defined by  $4000 < x < 4500 \ \mu m$ for  $y = 2500 \ \mu m$  in figure 9.10a and the line  $4000 < x < 4500 \ \mu m$  for  $y = 500 \ \mu m$ . The curvature towards the corner positions indicates that the latter are regions of high sensitivity. This contrasts with the 1-d case of figure 9.4, where the region furthest from the TESs was of highest sensitivity. Differences in form between the plots 9.10a and 9.10b are attributed to asymmetry in the TES configuration, an effect also apparent in the asymmetry of the peak in figure 9.9. Each TES is aligned with the y axis and is not located in the corner of the absorber (figure 9.7), and gives rise to marked differences in spatial resolution in the x direction ( $\Delta x$ ) compared to spatial resolution in the y direction ( $\Delta y$ ).

Since the response of each TES is non-linear with absorption position (figure 9.9) it follows that the spatial resolution will also vary across the surface of the absorber.

The position sensitivities in x and y are given by  $S_x = \frac{\partial \gamma_x}{\partial x}$  and  $S_y = \frac{\partial \gamma_y}{\partial y}$ , respectively and are plotted in figure 9.11(a, b).



Figure 9.11(a): Magnitude of the position sensitivity,  $S_x$  ( $\mu$ m<sup>-1</sup>) in the x direction.



**Figure 9.11(b)**: Magnitude of the position sensitivity,  $S_y$  ( $\mu$ m<sup>-1</sup>) in the y direction. Due to the asymmetry of the TES layout, the sensitivity in x is generally greater than in y.

The position sensitivity in x,  $S_x$  shows distinct minima at the mid-point between TESs (y = 2500 µm) at x  $\rightarrow$  0 µm and x  $\rightarrow$  5000 µm. This results from increased randomisation of phonon directions due to the greater probability of scattering as the absorber boundary is approached. In general,  $S_x$  is relatively uniform across the surface, with peak sensitivity close to the corner TESs. The sensitivity in y,  $S_y$  varies over the surface and the detector is less sensitive to displacements in this direction since the absorption point is tracking across the face of a given TES. The extent to which  $S_y$  is suppressed compared to  $S_x$  for this geometry depends on the size of the collecting area of each TES.

If a minimum value for the difference in  $\gamma$  for adjacent absorption positions is assumed, spatial resolutions,  $\Delta x$  and  $\Delta y$  may be deduced. Figure 9.12(a-d) shows  $\Delta x$  and  $\Delta y$  for  $|\gamma_1 - \gamma_2| = 0.01$ . This assumes that differences in the amplitude ratio of less than 1 % cannot be measured reliably. The absolute resolution may be improved if this condition is relaxed, but the profiles of the curves remain.

Figures 9.12(a, b) show that for most of the absorber surface, a spatial resolution in x,  $\Delta x \sim 100 \ \mu m$  is possible, assuming a minimum measured amplitude ratio difference of 1 %. The regions discussed previously close to the absorber boundary between the TESs at y = 2500  $\mu m$ , show the predicted poor spatial resolution (500  $\mu m$ ). These could be neglected by appropriate collimation of incident photons or by a modification to the detector configuration (section 9.5.3).



Figure 9.12(a): Spatial resolution,  $\Delta x$  (µm) shown in plan view.



Figure 9.12(b): Spatial resolution,  $\Delta x$  (µm) shown as a surface plot, truncated at  $\Delta x = 250$  µm for clarity.



Figure 9.12(c): Spatial resolution,  $\Delta y$  (µm) shown in plan view.



Figure 9.12(d): Spatial resolution,  $\Delta y$  (µm) shown as a surface plot.

From figures 9.12(c, d), a spatial resolution,  $\Delta y$  extrapolated from the sensitivity exceeds 1300 µm for mid x values. This is unacceptable for a detector intended for XEUS. The striking contrast between the  $\Delta x$  and  $\Delta y$  surfaces, and their complexity, is due to the arrangement of TESs. It may be concluded that the TES configuration is critical to the spatial performance of the detector (a conclusion also reached in the resistive node readout studies).

# 9.5.3 Square Bismuth Absorber with Corner TES Readout

To further investigate the effect of TES location on the spatial resolution and to achieve more symmetrical results in  $\Delta x$  and  $\Delta y$ , the model was modified to incorporate TESs which absorb phonons from the corners of the absorber only. In addition, the collecting area was reduced: instead of extending 10% of the distance along the y axis (500 µm), each TES now extends 3 % (or 150 µm) along both the x and y axis, as shown in figure 9.13. This has the effect of improving spatial resolution to the slight detriment of countrate, although pulse duration remains ~ 100 µs.



Figure 9.13: Configuration of the revised DROID model. The total collecting area of each TES is  $300 \ \mu m \ x \ 22 \ \mu m$ , a reduction of  $40 \ \%$  over the previous design.

The resultant spatial resolutions in x,  $\Delta x$  and in y,  $\Delta y$ , are shown in figure 9.14(a-d).



Figure 9.14(a): Spatial resolution,  $\Delta x$  (µm) shown in plan view.



Figure 9.14(b): Spatial resolution,  $\Delta x$  (µm) shown as a surface plot.



Figure 9.14(c): Spatial resolution,  $\Delta y$  (µm) shown in plan view.



**Figure 9.14(d)**: Spatial resolution,  $\Delta y$  ( $\mu$ m) shown as a surface plot. Asymmetries in these plots are due to statistical fluctuations in the model.

The most important point to note from inspection of these plots is that the response in x and y are now very similar with no regions of the absorber with spatial resolutions exceeding 250  $\mu$ m (the specification for XEUS). The improved uniformity would greatly simplify calibration of this detector. This has been achieved at the expense of  $\Delta x$ , which has been degraded from ~100  $\mu$ m to ~ 150  $\mu$ m as a result of the changes made to TES geometry. As expected, for both  $\Delta x$  and  $\Delta y$  the resolution is weakest close to the boundaries and between the TESs used for position resolution measurement (i.e.

TES 1 and TES 2 for determination of  $\Delta x$ ). In a detector, incident X-rays are collimated to eliminate stray photon interaction with the TESs and substrate. If the collimation restricts the photons to the central regions of the absorber, this effect is negated.

# 9.6 Conclusion

To conclude, this model demonstrates that it is possible to determine X-ray absorption position in bismuth at 100 mK by the analysis of pulse amplitudes. The spatial resolution may be improved by the inclusion of additional information regarding either the peak arrival time or the extrapolated leading edge of the pulse. For fast pulses (small, thick bismuth absorbers or those with a copper layer), the magnitude of this improvement may be limited by the finite response time of the readout electronics. Since the modelled TES pulses indicate that pulse amplitude is highly sensitive to position, resolution by pulse timing has not been pursued here. The model highlights the importance of the TES layout on the uniformity of spatial response, and demonstrates that spatial resolutions better than 250  $\mu$ m are possible for a bismuth absorber of side length 5 mm operating at 100 mK, with a symmetrical TES configuration.

# **Chapter 10**

# **Conclusions and future prospects**

#### **10.1 Conclusions**

This thesis has described the early stages of development of a new technology for X-ray detection. As stated in Chapter 1, other groups around the world are pursuing similar goals (NIST, Colorado [Wollman, D., 2000]; SRON, Utrecht [Hoevers, H., 1999b]; Goddard Space Flight Center [Stahle, C., 2000a]). However, a stable TES-based X-ray microcalorimeter optimised for X-ray astronomy is yet to be demonstrated. Our approach differs from that of other groups by placing greater emphasis on understanding the physics behind the materials and operation of cryogenic microcalorimeters, with a long-term view to produce stable, well-optimised detectors. During the course of this work, the impact of materials science on the design of detectors has become apparent to the international community. Ageing of the TES thermometer due to diffusion and alloy formation processes has proven to be a major obstacle to the development of stable devices. Our published results (Holland, A. [1999], and described in Chapter 6) regarding the susceptibility of Al/Ag devices to degrade was the first publication on this topic and have been cited as justification for the investigation of new TES material combinations [e.g. Fukuda, D., 2000]. Our subsequent investigations into Al/Ag and Ti/Au bilayers and Ag/Al/Ag and Cu/Nb/Cu trilayers were also described in Chapter 6. The conclusions drawn from studies of changes in transition temperature and room temperature resistance with time, and from analysis of Auger electron spectroscopy of all of these material combinations, have directed our design of subsequent detectors towards TESs fabricated solely from iridium. In Chapter 7, initial superconducting results from cooling a series of test iridium structures were presented. We have demonstrated a degree of control over the transition temperature by correlation of  $T_c$  with deposition parameters, notably substrate temperature and layer thickness. A tentative (pending further data) physical model for this correlation is proposed, based on the growth of an iridium silicide proximity layer during deposition. The model indicates that an iridium silicide layer of ~ 3 nm is sufficient to reduce the superconducting - to - normal transition temperature from 289 mK to 52 mK. In summary, the convergence of results in favour of iridium TESs and our growing understanding of the interaction of iridium with other materials in the detector gives confidence in the stability and potential performance of these devices.

Some of the key constraints upon the design of a TES are outlined in Chapter 5. Identification of these constraints enables designs to be optimised for a range of applications. For example, there are trade-offs in the design between maximum count rate (which requires high thermal conductance between

electrons and phonons in the TES, and hence a large TES volume) and the requirement for no superconducting/normal phase separation, which limits the maximum TES length. Many of these parameters are material dependent. Chapter 5 also emphasised the inclusion of test structures on our detector chips for materials analysis and further investigation of the superconducting properties of iridium, as this will form a large part of our future work.

Much of Chapter 3 was devoted to a description of the liquid helium dilution refrigerator and the use of cryogenic techniques in the laboratory. The TES detector programme is the first project at Leicester in the area of cryogenic X-ray detectors and our current knowledge of dilution refrigerator operation has be gained entirely during the course of this work. The physics at low temperatures (and consequential engineering challenges) has been a new field for our group, with application beyond TES detectors, most notably in the investigation of micro channel plate resistance at liquid helium temperatures (Chapter 4). Experimental techniques were further explored in Chapter 8, which includes the results of X-ray testing of a single pixel TES with our new digital signal processing chain. We were able to demonstrate that this was capable of reproducing the energy resolution measurement (25 eV FWHM at 6 keV) of an equivalent analogue processing chain, and was also a valuable tool in the investigation of anomalous spectral features.

Astronomy applications demand imaging capability. One means of achieving this is to use extended absorbers with multiple TES output. This route was examined in Chapter 9 with particular emphasis on bismuth absorbers. We have pursued the extended absorber approach in contrast to many other groups' preference for pixellated detectors, with the belief that it offers the best compromise between spatial resolution and cost of development. Interestingly, Goddard Space Flight Center have recently published a paper on initial designs of a very similar concept [Figuero-Feliciano, E., 2000] and interest seems to be growing in general for detector configurations that do not require large multiplex SQUID systems, since this continues to be a difficult problem to overcome. The results of modelling heat flow through linear bismuth absorbers were presented. Analysis indicates that despite the poor thermal conductivity of bismuth, it is possible to fabricate a detector (from multiple 'strip' detectors, each terminated with a TES) that meets the spatial resolution requirements of XEUS (~  $250 \,\mu$ m).

#### **10.2 Future Prospects**

Progress has been rapid in the development of cryogenic detectors. This progress looks set to continue with the maturity of a number of novel pixel array and distributed readout imaging detector concepts (Deiker, S. [2000]; Figueroa-Feliciano, E., [2000]; Bruijn, M. [2000b]). With our industrial partners, Oxford Instruments, we are well placed to pursue an imaging TES detector. However, much further work remains to be done, notably in the characterisation of the extended bismuth absorber design. Testing with heat pulses to investigate the phonon distribution and flow pattern will enable us to constrain some of the design parameters in order to proceed to X-ray testing of the optimised detector.

This is expected to form the major focus of our activities over the next two years. In parallel with these experiments, X-ray testing of single pixel detectors will be used to determine our system noise and investigate methods of reducing this noise. The time required for such investigations should not be underestimated. Additionally, our research into the stability and degradation mechanisms of TES materials will continue with use of facilities both at Loughborough University and Cambridge University, within the framework of the UK X-ray Cryogenic Spectrometer Programme (XCSP). A full understanding of the active degradation processes is essential for the successful deployment of a space detector, for reasons stated in Chapters 5 and 6. We also intend to feedback SQUID results from X-ray testing to Oxford Instruments to facilitate their Super-SQUID development programme. Regardless of the final configuration of the detector, SQUIDs will be an integral part of the device and need to be included in the design iterations from an early stage of development.

In summary, the results presented in this thesis form the basis for a thorough understanding of the physics of TES X-ray detectors which will enable us to develop a high-energy and high-spatial resolution microcalorimeter for use in astronomy and ground-based X-ray microanalysis spectroscopy.
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