TEMPERATURE CONTROL INSTRUMENTATION FOR
SCANNING TUNNELLING MICROSCOPY

by

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Abstract

This thesis describes three different design projects that are intellectually connected by the fact that they all involve the development of apparatus to facilitate the precise control of sample temperature in modern microscopes.

The first project is a low-temperature sample stage, for a beetle-type scanning tunnelling microscope. The design for this sample stage, and images taken on it with atomic resolution at 114 K are presented. This stage has the capability for variable-temperature sample cooling, which is also discussed.

The second project is a set of low- and variable-temperature isothermal radiation shields for a new microscope that is currently being designed and assembled by our research group. These shields provide temperature control between 5 K and room temperature, with measured stability better than ±0.1 K. Controlled and stable temperature changes at rates up to 1.5 K per minute have been produced. The shields are modular and can easily accommodate future modifications. The design for the shields, along with their cooling and temperature control capabilities, is presented.

The third project is a new stage design for heating, cleaning, and transferring metal and semiconductor samples. Also for use with the new microscope, this stage uses electron bombardment to provide precision temperature control between room
temperature to temperatures in excess of 1250 °C. With this stage, the sample temperature can be determined by measuring the power applied to the sample. The design of this stage, its heating performance, and a method to calculate the sample temperature is presented.
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Table of Contents

Abstract i

Acknowledgements iii

Table of Contents iv

List of Tables vii

List of Figures viii

Chapter 1:

Introduction 1

1.1 Scanning Tunnelling Microscopy 1

1.2 Motivation 4

Chapter 2:

Literature Review 8

2.1 The Development of Low- and Variable-Temperature STM 8

2.2 Low- and Variable-Temperature STM Instrumentation Review 12

Chapter 3:

Variable-Temperature STM Sample Stage 24
3.1 Introduction .............................................. 24
3.2 Design Goals ............................................ 27
3.3 Description of the Cold Stage ....................... 27
3.4 Thermal Modelling ..................................... 38
3.5 Testing .................................................. 45
3.6 Recommendations ...................................... 54

Chapter 4:

Isothermal STM Cold Cans ................................. 57
4.1 Introduction ............................................ 57
4.2 Design Goals And Requirements .................... 58
4.3 Description of the Cold Cans ......................... 59
4.4 Testing .................................................. 61
4.5 Recommendations ...................................... 70

Chapter 5:

The Sample Preparation Stage ............................. 71
5.1 Design Goals and Requirements ....................... 72
5.2 Description of the Sample Preparation Stage ......... 72
5.3 Testing .................................................. 77
5.4 Summary ............................................... 84
5.5 Recommendations ...................................... 84

Chapter 6:

Conclusion .................................................... 85
Bibliography .................................................. 87

Appendix A:

Chamber Design for the Isothermal STM ............ 92
List of Tables

3.1 Contact resistance estimates from look-up table. . . . . . . . . . . . . 41
3.2 Contact resistance values. . . . . . . . . . . . . . . . . . . . . . . . . 42
3.3 The results of thermal modelling. . . . . . . . . . . . . . . . . . . . . 45
4.1 Liquid nitrogen cooling results summary. . . . . . . . . . . . . . . . . 67
4.2 Liquid helium cooling results. . . . . . . . . . . . . . . . . . . . . . . 68
5.1 Curve fitting results. . . . . . . . . . . . . . . . . . . . . . . . . . . . 81
List of Figures

1.1 A photograph of the beetle-type STM (B1). ........................................... 2
1.2 The first results published by Binnig and Rohrer. ................................. 2
1.3 The original Si(111)-7\times7 recording. ............................................... 4

2.1 A rendered image of “IBM” spelled out with Xe atoms. ....................... 15
2.2 A schematic of Meyer’s STM. ............................................................... 17
2.3 A schematic of the STM designed by Behler et al. ............................... 20
2.4 A schematic of Stipe’s STM ................................................................. 23

3.1 A filled-state image of a Si(111)-Ge(5x5). ......................................... 26
3.2 An image of the cold stage mark 1 ......................................................... 28
3.3 A plot of radiative heat transfer rate. .................................................... 31
3.4 A simplified “section view” of the cold stage ......................................... 33
3.5 A top view schematic of the cold stage. ............................................... 35
3.6 An image of the cold stage mark 2. ....................................................... 37
3.7 A schematic of the thermal model components. .................................... 39
3.8 Cooling curve plot of the thermal model using the “table” contact resistances ................................................................. 43
3.9 Cooling curve plot of the thermal model using the “equation” contact resistances ................................................................. 44
3.10 Cooling curve plot of actual data from the mark 2. .......... 48
3.11 A plot showing the fit to the cooling data. .................. 50
3.12 Images taken on the cold stage demonstrating the drift observed while cooling. ........................................ 51
3.13 An image of Si(111)-7×7 taken on the cold stage at ~114 K. .... 53

4.1 Four images of the cold cans during assembly. ................. 60
4.2 A schematic of the cooling system. ............................. 62
4.3 A plot showing liquid nitrogen cooling data for the cold cans .... 66
4.4 A plot showing liquid helium cooling data for the cold cans ....... 69

5.1 The main body of the Sample Preparation Stage. .............. 73
5.2 The sample preparation stage assembled ........................ 74
5.3 The “wire bundle” for the SPS .................................... 75
5.4 The sample preparation stage assembled with a sample ........ 76
5.5 An image of the modular electron-beam heating plate. ........ 78
5.6 A molybdenum sample plate being heated. ................. 80
5.7 A plot of sample temperature and power for the SPS. ........ 82
5.8 A plot of the curve fitting results ............................... 83

A.1 A rendered image of the ISO system. .......................... 93
A.2 An image showing the SPS in the prep chamber. ............. 93
A.3 An image of the entire isothermal system. .................... 95
Chapter 1

Introduction

This chapter provides a brief historical look at the development of scanning tunnelling microscopy (STM) and introduces the work presented in the remainder of this thesis. The next chapter will delve more deeply into the literature and background behind low- and variable-temperature instrumentation for STM.

1.1 Scanning Tunnelling Microscopy

Scanning tunnelling microscopy is a powerful local probe technique for investigating conductive surfaces at the atomic level. It can also be applied to adsorbates on these surfaces, such as atomic and molecular species, and thin insulating layers. Real-space topographic images of the electronic local density of states of a conductive surface can be acquired with atomic resolution. Information about the electronic structure of atoms and molecules can also be acquired using scanning tunnelling spectroscopy.

The physical basis for STM is quantum mechanical tunnelling. It was predicted by planar tunnel barrier theory [10] that the resistance of the tunnelling gap, $R(s)$,
Figure 1.1: This photograph shows the beetle-type STM (B1) designed by Moffat and McLean [21]. This beetle has been used by students in McLean’s research group since 1999. It is shown here on top of the cold stage, that is described in chapter 3 of this thesis.

Figure 1.2: These are the results presented by Binnig and Rohrer’s in their first publication [5]. The topography of the CaIrSn₄ surface resolving mono-atomic steps is shown in 1.2a. A micrograph of a gently corrugated Au(110) surface with one four-atomic layer step is shown in 1.2b. Reprinted figure with permission from [5]. Copyright 1982 by the American Physical Society.
CHAPTER 1. INTRODUCTION

would have the following dependence on the tip-to-sample distance,

\[ R(s) \propto \exp(C\phi^{1/2} s), \]  

(1.1)

where \( C \) is a constant, \( \phi \) is the average barrier height of the gap, and \( s \) is the gap width between the tip and the sample. Tunnelling through a vacuum barrier with control over gap distance was first experimentally demonstrated in 1981 by Binning and Rohrer [3]. Their junction consisted of a fixed platinum plate and a tungsten tip on a mobile platform. With this apparatus they observed the predicted exponential dependence of tunnelling resistance on the width of the gap. Measuring the vacuum tunnelling current was a very important step towards the realisation of a microscope. Then, in 1982, they published the first images using their newly developed technique, scanning tunnelling microscopy [5]. In this publication they presented topographic images, acquired by their STM, of mono-atomic height steps on an otherwise flat CaIrSn\(_4\) surface (figure 1.2a) and of a Au(111) surface at room temperature and 300 °C (figure 1.2b). Initially, many people were sceptical of these results, as the resolution obtained could not be fully explained. Some also deemed that the technique was too experimentally demanding for wider adaptation outside of the IBM laboratories. Much work was still needed to convince the general surface science community to accept the STM [2].

The result that perhaps did the most to advance the cause of the STM was the image, shown here in figure 1.3, of the Si(111)-7\( \times \)7 reconstructed surface [4]. At that time, the Si(111)-7\( \times \)7 reconstruction was one of the most intriguing unsolved problems in surface science. Real-space images, provided by the STM, of the 12 adatoms in the Si(111)-7\( \times \)7 unit cell provided new information and were a big step towards knowing the complete structure of the reconstruction. In fact, these results
excluded all of the existing theories concerning the Si(111)-7×7 reconstruction. It wasn’t until 1985 that, based on transmission electron diffraction and microscopy, the currently accepted model for the Si(111)-7×7 reconstruction was published [33]. Even so, these images grabbed the attention of the surface science community, proving to it that STM was a useful technique [2]. From that time forward, STM underwent rapid development as many research groups adopted and adapted it for their own applications [12, 24]. In 1986, Binnig and Rohrer were awarded the Nobel prize for the invention of the scanning tunnelling microscope.

### 1.2 Motivation

This thesis describes the development of two different variable temperature apparatuses that will allow samples to be imaged with a scanning tunnelling microscope.
below room temperature. It also describes the development of a new sample heating stage that allows the sample to be heated controllable to temperature in excess of 1250 °C. All of these projects involve controlling the temperature of a sample in microscopy systems.

For microscopy, controlling the sample temperature offers a number of benefits over simply operating at room temperature. The tip-sample junction is extremely sensitive to differential thermal expansion. Allowing the sample and microscope temperature to vary with the ambient room temperature is not ideal. By controlling the sample temperature with a cryostat, the temperature is determined by the user and is less prone to random variations. Already in their first publication about vacuum tunnelling [3], Binnig and Rohrer commented that operating at low temperatures would offer “better mechanical stability” because “thermal stress release and thermal expansion become less bothersome”. As temperature decreases, thermal expansion coefficients decrease as well [34]. This means that temperature variations at low temperatures result in smaller expansions and contractions than at room temperature. Temperature control also opens up the possibility of studying phenomena, such as phase transitions or adatom diffusion, that are either difficult or impossible to perform at room temperature. As such, sample temperature control is a feature included as part of many microscopy systems. Unfortunately, there are a number of difficulties that come with introducing cooling which prevent it from being standard in all microscopy systems. Cryostats are a significant source of mechanical noise, which can easily produce too much noise for a microscope to operate well.

The 3rd and 4th chapters of this thesis will each present the development and performance of different cooling solutions. The cold stage is a low temperature sample
stage that upgraded a beetle-type microscope. It has allowed low temperature experiments to be performed for the first time within our research group. Operating with liquid nitrogen, it has a minimum sample temperature of 114 K. The cold cans were designed second. They will be used with the new microscopy system being assembled, that will operate temperatures down to 4 K. Temperature control between room temperature and base temperature with the cold cans has been demonstrated with precision better than ± 0.1 K.

Sample cleaning and preparation is a surprisingly delicate process that requires a high degree of temperature control. When a bulk crystal is cleaved, and a new surface is exposed, the configuration of the atoms at the surface is very rarely the same as in the bulk. Immediately after being exposed, surface atoms attempt to find the lowest possible energy state. These atoms will often move away from their bulk positions, reconfiguring themselves at the surface. This is referred to as a surface reconstruction. The Si(111)-7×7 surface is probably the most well known surface reconstruction. To prepare a clean 7×7 surface, the typical procedure involves flashing the Si(111) sample to 1250 °C and then annealing at 850 °C. The flash evaporates off the first few atomic layers of Si, and hopefully any contaminants also evaporate or alternatively diffuse into the bulk. The longer anneal gives surface atoms time to find their lowest energy state and form a well ordered surface. To ensure that the desired surface can be prepared consistently, it should be possible to heat the surfaces to high temperature and also control the temperature precisely.

The sample preparation stage (SPS) presented in the 5th chapter of this thesis was designed to provide the temperature control and high temperatures necessary for both sample cleaning and preparation. Like the cold cans, it will be used in the new
microscopy system being designed by Drevniok, McLean, and Visser. This system was designed to work with both metal and semiconductor samples. Previous heating stages have resistively heated the sample for flashing and annealing. This was feasible because they were designed for use with mostly semiconductor sample. Resistive heating would not work with metal samples. Instead, the SPS heats samples by electron bombardment. In electron bombardment, or electron beam heating, electrons from a hot filament are accelerated across a vacuum gap towards a biased sample. This method can be used to heat both metal and semiconductor samples. It will be demonstrated that the sample temperature can also be inferred from the input power using a calibration curve.
Chapter 2

Literature Review

In this chapter, the first stages in the development of low- and variable-temperature instrumentation for STM will be outlined. A number of past and present instruments will be described and categorised.

2.1 The Development of Low- and Variable-Temperature Scanning Tunnelling Microscopy

Imaging resolution is one of the foremost concerns when performing scanning tunnelling microscopy. In order to obtain atomic resolution, the vibrational noise at the tunnelling junction must be less than $\sim 1$ pm, significantly less than the size of an atom. Otherwise, the signal from individual atoms will be indistinguishable from the noise. In their first paper, Binnig and Rohrer predicted that lowering the temperature of the microscope will have a positive affect on mechanical stability [5]. They expected this result because “thermal stress release and thermal expansion become
less bothersome” at low temperatures.

Binnig and Rohrer’s first microscope had liquid helium cooling and super conducting magnetic levitation. Though they admitted that it was too complicated and was never successfully operated [2], it was the first low-temperature scanning tunnelling microscope. The challenges of operating at low-temperature were significant, as it wasn’t until 1985 that another low-temperature microscope was reported. Drake et al. designed a microscope to operate in air, oil, and liquid nitrogen [8]. They used a bath-type cryostat, where an inner and an outer dewar are filled with liquid nitrogen. A mechanical pump evacuated the inner dewar where the microscope was located to remove as much water vapour as possible. Noise from the boiling of the liquid nitrogen was reduced with boiling chips. The microscope was moved into and out of the cryogen bath with a motor. The volume of cryogen required to fill the dewars was not mentioned, but with full dewars, 16 hours of operating time was available. While operating with liquid nitrogen cooling, they were able to image atoms on a $2\text{H-TaSe}_2$ surface. At room temperature, they were not able to attain atomic resolution while operating under either air or paraffin oil.

In 1986, Smith and Binnig published a design [27] for an ultrasmall scanning tunnelling microscope. Their design was motivated by the desire to increase resonant frequencies in the microscope and to fit the microscope into a “standard-sized” liquid helium dewar. With exception to the never-used original from Binnig and Rohrer, this was the first reported scanning tunnelling microscope to operate at liquid helium temperature. There was nothing special about their cooling system, as it was simply a liquid helium dewar. The microscope fits into a 35 mm diameter tube, which was inserted into the dewar to rapidly cool the microscope. This microscope was used
to observe the phonon spectrum of graphite [28]. Though no images taken with this microscope could be found in literature, the authors reported imaging the atomic structure of graphite.

A few months later, another design for a low-temperature scanning tunnelling microscope was published [18]. This design was unique in that it operated in an ultra-high vacuum environment at liquid helium temperatures. The cooling system consisted of copper sample holder cooled by direct contact with a liquid helium dewar. The sample stage was surrounded by two heat shields held at 4 and 77 K. Both topographic and spectroscopic results are reported for NbN and graphite surfaces. For these measurements, the microscope was operated at 6.8 K. Very low drift rates were observed. At 6.8 K, lateral drift was less than 2 nm/h and vertical drift was less than 0.65 nm/h. No information was given on consumption rate of the liquid helium.

All of the low-temperature microscopes reported up until this point have operated at fixed temperatures. Typically operating at either room temperature or the temperature of the cryogen. Perhaps the most famous scanning tunnelling microscopy experiment was performed with such a microscope. This was the work of Eigler and Schweizer, and in particular, their image of single Xe atoms forming the letters “IBM” on a Ni(110) surface [9]. For this work they used a microscope that operated at 4 K. Unfortunately, little else is reported about the microscope.

An important development occurred in 1992, when Wolkow presented [36] a new design for a variable-temperature scanning tunnelling microscope. This microscope could operate between approximately 80 to 350 K in ultra-high vacuum. Variable-temperature operation was very useful and allowed investigation into new phenomena
Diffusion of adsorbates, phase transitions, and other temperature dependent effects can all be more readily studied with a variable-temperature instrument. One of the critical features of this design was the cryostat. Instead of using a bath or dewar, as in the previous systems, this design used a continuous flow helium cryostat. It was found to generate less mechanical noise than a liquid nitrogen bath cryostat. By adjusting the rate of cryogen flow through the cryostat, the temperature of the microscope and sample was controlled. A single heat shield protected the microscope against radiative heating. The shield was connected to the cryostat for cooling, but openings in the shield for sample and tip transfer reduced its effectiveness. To provide the necessary mechanical decoupling, the microscope was suspended by springs from the cryostat. The thermal connection through the springs was deemed insignificant. The electrical connections to the microscope were made with 40-gauge, single-conductor copper wires. These wires provided an important thermal connection between the microscope and the cryostat. During operation, the microscope was suspended and the thermal connection through the wires helped to maintain the desired temperature. A clamping mechanism pushed the microscope against the cryostat and was used when rapid temperature changes were needed. In this design, it was reported to take 30 minutes to cool the microscope from 295 to 100 K. Once the temperature was stable, thermal drift was found to be less than 0.1 Å. A chosen temperature could be maintained for over 10 h to within 2 K. To demonstrate the performance of his new design, Wolkow imaged the buckled dimers on the Si(001) surface at 120 K [35, 36].
2.2 Low- and Variable-Temperature Tunnelling Microscopy Instrumentation Review

In this section, a review of low- and variable-temperature cooling systems for tunnelling microscopy systems will be presented and the instruments will be classified by core characteristics. A variety of tunnelling microscope designs with sample temperature control have been presented in scientific publications since the invention of the STM. In fact, sample temperature control is now common for most new microscopes. Temperature control has been achieved through many different methods and there is no generally accepted “best” solution or design. Each approach has its strengths and weaknesses.

To classify the different cooling apparatuses, a few key design characteristics were identified. First, the source of cooling power was usually either a bath cryostat or an open-flow cryostat. Bath cryostats typically have lower cryogen consumption rates, as well as lower noise levels, when compared to open flow cryostats. Open flow cryostats offer the capability for easier variable temperature operation and faster temperature changes. With flow cryostats, the temperature can be controlled by adjusting the cryogen flow, and additionally, heating does not cause additional expenditure of the cryogen. For these reasons, open flow cryostats have been chosen for well controlled variable-temperature operation.

A few microscopes that used bath cryostats, also allowed variable-temperature operation. So, the bath cryostat systems have been divided into those that operate at fixed temperatures and those that are variable-temperature.

The final division was made by looking at whether the cooling was isothermal or
Isothermal cooling was performed to the entire microscope and sample, while the alternative was to cool only the sample, while the microscope remained close to room temperature. Isothermal operation benefited by having lower thermal drift, as thermal expansion coefficients were reduced at cryogenic temperatures. Unfortunately, as mentioned in the previous chapter, the scan range of the microscope is also reduced at cryogenic temperatures. By not cooling the microscope itself, the full scan range is available at any sample temperature. Non-isothermal cooling systems also typically have a smaller mass undergoing temperature changes, which results in faster temperature changes.

A number of other characteristics can be used to further categorise the designs, but the three just mentioned were the main defining characteristics. These other characteristics were smaller features, incorporated to increase the cooling or imaging performance of the microscope.

Typical performance metrics for temperature control stages include the base sample temperature, the time required to cool the sample to base temperature, the cryogen consumption rate, the drift between the scanner tip and the sample, the sample temperature range accessible, and ultimately, the “quality” of the images taken with the microscope. Image quality is rather subjective, but the ability to clearly resolve individual atoms is typically the baseline for tunnelling microscope images.

### 2.2.1 Fixed Temperature, Isothermal Bath Cryostat Designs

Bath-type cooling systems were the first to be used. One of the well known low-temperature microscopes that has used a bath-type cryostat was designed by Gaisch et al. in collaboration with Schneider. Described in a 1992 publication [11], the
cooling system used a custom designed helium-bath cryostat to isothermally cool the microscope, the sample, and the inner shielding. Outside the helium shield was a shield cooled by a liquid nitrogen dewar. A minimum sample temperature of 8.5 K is reached in approximately 3 hours. This microscope operates at either room temperature or its base temperature. It has no capability for temperature control at any other temperature. The capacity of the liquid helium dewar was not mentioned, but it was empty after operating for approximately 10 hours.

Eigler’s low-temperature microscope, was used for some of the most famous and important STM experiments done to date, including the first demonstration of positioning single atoms [9]. This microscope operated at a fixed temperature of 4 K. In this experiment, Eigler and Schweizer moved xenon atoms on a Ni(110) surface to form the letters “IBM”, as seen in figure 2.1. What was perhaps most unique about the design was the use of a pendulum for vibration isolation. This allowed the microscope to swing freely at approximately 1 Hz [23, 13]. The pendulum with the microscope hangs from stainless-steel bellows inside a helium gas exchange canister. The canister is inserted directly into a liquid helium dewar. After evacuation, the canister is back-filled with helium gas to provide thermal coupling between the microscope and the liquid helium bath. The entire microscope sits at 4 K and consumes 0.5 Lh$^{-1}$ of liquid helium.

2.2.2 Variable Temperature, Isothermal Bath Cryostat Designs

A design for a variable temperature microscope was presented by Smith and Shih in 1995 [26]. This microscope was made for the purpose of studying low-dimensional
quantum heterostructures. To facilitate these studies, a precision two-dimensional translation stage was needed. The translation stage was necessary to position the microscope tip within the epilayer region that contained the heterostructures being studied. The cryostat consisted of a two-part cryogen reservoir, a removable liquid nitrogen jacket, and an actuated aluminium cold shroud. The two-part reservoir could be filled with either liquid helium or liquid nitrogen. The upper and lower parts of the reservoir were inside the vacuum chamber and were connected by fill tubes that passed through an 8” Conflat (CF) flange. The cold shroud could be externally actuated to allow more or less radiation to fall on the microscope. By raising or lowering the cold shroud to different positions, temperatures between base and room temperature could be attained with stability. In its lowest position, the cold shroud was pressed firmly against the cryogen reservoir for cooling. During liquid helium operation, a liquid nitrogen cooled jacket shielded the microscope and two-part reservoir. This jacket could be removed to simplify operation of the microscope when liquid nitrogen was used in the reservoir. The microscope was suspended from the flange by weak springs. These springs, along with eddy current damping, reduced the noise from the
boiling liquid nitrogen and from other sources outside of the chamber. For testing, they removed the jacket and used liquid nitrogen. In this configuration, the sample temperature reaches 120 K in 3 hours, and 80 K with an additional 3-5 hours. They reported that imaging with atomic resolution has been performed at temperatures down to 77 K, though images in the articles only show surfaces down to 203 K. In a later publication, they show images of a Si(001) surface at 127 K [25], but this was the lowest temperature found.

The design presented by Meyer in 1996 was for another bath-type cooling system [20]. This variable-temperature instrument operated in the range of 15 to 300 K. Different from the Smith and Shih design, this system used a small resistance heater placed by the sample to provide temperature control. Two radiation shields surrounded the microscope as shown in figure 2.2. The inner copper shield was held at 4 K by the helium bath, while the outer aluminium shield sits around 100-140 K and is cooled by the cryostat’s radiation shield. For vibration isolation, the microscope hangs from springs and has eddy current damping. When hanging free, the thermal connection to the helium bath is very weak and the microscope experiences heating through the electrical wires. For cooling, a wobble stick was used to push the microscope against the inner radiation shield. From room temperature, it takes 4 to 5 hours for the microscope to reach 30 K. To reach lower temperatures, a braid could be installed between the inner shield and the microscope. This noticeably increased the noise levels, but also brought the sample temperature down to 15 K. With the braid, the sample was fixed to 15 K. The typical liquid helium consumption rate was 0.3 Lh$^{-1}$. The performance of the microscope was demonstrated by imaging a Ag(111) film at 80 K, laterally manipulating individual CO molecules on a Cu(211)
surface at 15 K, and vertically manipulating individual Xe atoms, also on a Cu(211) surface at 15 K. Images of the atomic scale manipulation are shown in figure 2.2. They estimated thermal drift to be less than 0.1 nm/h at 15 K.

The “Eigler-style” microscope was improved upon in 2001, when Rust et al. published an article on how to add the capability for variable-temperature operation to this microscope. They found that by adjusting the pressure of the helium gas in the exchange canister and counter-heating with a wire-wound resistor, stable temperature control between 4 K and 60 K was possible. Helium consumption could not be reduced without causing the temperature to become unstable.
2.2.3 Variable Temperature, non-Isothermal Flow Cryostat Designs

In 1994, Horch et al., from Comsa’s research group, published an article describing their variable-temperature microscope [14]. They wanted to be able to investigate temperature dependent effects, and for this they needed precision temperature control over a wide range of temperatures. To achieve this, they used a helium flow cryostat. A soft copper braid connected the cryostat to the sample holder. By using a braid, the competing requirements for vibrational decoupling and good heat transfer were satisfied. Rapid temperature changes were possible, in part, because the mass being acted upon was kept very small. Accordingly, this was a non-isothermal design with only the sample holder and the sample experiencing direct heating or cooling. The microscope itself and other supporting structures were left at room temperature. With this instrument, sample temperatures between 10 and 400 K were accessible. Cooling the sample from 300 to 10 K took only 6 minutes.

Less than a year later, another variable temperature microscope was presented by the same group [6]. This design had many features in common with the last one, but also boasts a number of improvements that have increased the upper temperature limit and reduced noise levels. The braid connecting the cryostat to the sample holder was found to transmit significant vibrations. Vibrational noise was measured quantitatively by a fast Fourier transform [FFT] of the tunnelling current. First, the resonant modes of the cryostat were increased by fixing the cryostat to rigid tubes running parallel to it. Second, noise in the braid was damped by clamping it to a large mass part way along its length. Pushing and pulling screws joined the braid to the large mass, providing a rigid and thermally isolating connection. These two
modifications reduced the noise in the tunnelling current such that it was almost undetectable in the FFT. Clamping the braid had the secondary effect of raising the base sample temperature by 5 K. Sample temperatures can now range between 20 K and 700 K. With this microscope, densely packed metallic surfaces have been imaged with atomic resolution at temperatures down to 20 K. Using liquid nitrogen, samples can be cooled from room temperature to 100 K in 25 minutes.

In 1997, a new continuous flow cryostat cooling system for variable-temperature tunnelling microscopy was published by Behler et al. [1]. This system was developed independently of those from Comsa’s group [6, 14], but shared many similar design concepts. The limitation of most previous instruments, most using bath-type cryostats, was that they could not perform rapid temperature changes. Behler et al. desired to be able to change temperature at a rate of tens of degrees Kelvin per minute. As before, the solution was to reduce the mass that was being cooled. This naturally leaded to having a non-isothermal cooling system, where only the sample was cooled. A specially designed continuous flow cryostat provided the necessary cooling and produced a minimal level of noise from helium vibrations. Interestingly, by tilting their cooling system 2.5° away from horizontal, the base temperature of the cryostat was reduced from 8 to 6 K. They explain this by saying that when the cryostat was level, there was a convection current within the helium, that was reduced by tilting the cryostat. A soft copper braid connected the cryostat to the sample holder. Silicone O-rings provide the necessary vibrational damping between the microscope and the chamber. Figure 2.3 shows a schematic of their entire cryostat and microscope. The sample temperatures accessible range from 20 to 300 K. Cooling to the base sample temperature of 20 K takes approximately 60 minutes. With a tungsten
filament close to the sample, the sample could be heated radiatively and cleaned by flashing up to 500 K for a few seconds. After flashing, the sample returns to 20 K in approximately 3 minutes. By adjusting the helium flow rate, the microscope could be operated at temperatures up to 80 K. While the tungsten filament could be used to actively heat the sample, it was found to cause excess outgassing from surrounding surfaces as they heated. For variable temperature operation, the sample was cooled to 20 K, and then helium flow would be reduced. Temperatures would typically take 30 minutes to stabilise.

The Center for Atomic-scale Materials Physics [CAMP] research group at the University of Århus in Denmark designed a remarkable variable-temperature microscope...
This microscope has been used to take “STM movies”\(^1\); that is, consecutive images of the same area on a surface at rates up to 1 image/s. This is a non-isothermal microscope that uses a soft copper braid to create a permanent between the continuous flow helium cryostat and the sample stage. The flow cryostat used was the one designed and used by Comsa’s research group [6, 14]. Using a flow cryostat and a braid allowed for fast temperature changes. The entire microscope assembly and a secondary decoupling mass were suspended by Viton rings. The braid was clamped to the secondary mass to damp vibrations transmitted through the braid to the microscope [6]. A Zener diode mounted close to the microscope provided counter-heating. Using the diode, the microscope itself could be kept at a constant 285 K for all sample temperatures. The scanner is isolated from the sample holder by three quartz balls. An annealing filament was located close to the sample to provide stable temperature control above the base temperature. Using liquid helium, the sample was cooled to approximately 25 K within 30 minutes. Using the annealing filament, the sample temperature could be controlled between 25 and 120 K. In this temperature range, the cryostat was kept at 4 K. For higher temperatures, the cryostat temperature was increased by reducing the helium flow. For these higher temperatures, liquid nitrogen could be used in the cryostat instead of liquid helium. With liquid nitrogen, the sample reached 116 K in approximately 60 minutes. By operating with liquid nitrogen and using the annealing filament, the sample temperature could be controlled between 116 K and room temperature. Atomic resolution was routinely obtained at low temperatures and at room temperature.

\(^1\)http://www.phys.au.dk/spm/stmmovies.shtml
2.2.4 A Variable Temperature, Isothermal Flow Cryostat Design

Following the example first set by Wolkow, Stipe et al. published the design of their continuous flow cryostat based variable-temperature microscope in 1999 [31]. This design seems to have combined many of the best features of existing microscopes. The cooling system design was very similar to Meyer’s, with inner and outer radiations shields; the inner shield was bolted directly to the cryostat, and the outer shield was cooled by the exhaust from the cryostat. For vibration isolation, the microscope was suspended from weak springs in the inner can and used eddy current damping. Rapid cooling and temperature changes were facilitated by clamping the microscope against the inner can with a screw. Sapphire windows in the cans allowed for light access to the tip-sample junction. Physical access for sample and tip exchange was provided by doors in the cans. All electrical wires running to the microscope were heat sunk to the inner shield via a sapphire plate. A detailed schematic of the microscope and also the entire system can be seen in figure 2.4. This microscope was shown to be very stable with drift in the tip-sample space of 0.1 pm/min and could image the same area of a surface while the temperature was changed from 80 to 8 K.
Figure 2.4: A schematic drawing of Stipe’s variable-temperature microscope, as well as, an overview of the entire system [31]. Reprinted with permission from [31]. Copyright 1999, American Institute of Physics
Chapter 3

Variable-Temperature Scanning Tunnelling Microscopy Sample Stage

3.1 Introduction

In this chapter, the design and performance of a variable-temperature sample stage (hereafter called the “cold stage”) will be presented. The cold stage has allowed tunnelling microscopy to be performed on samples below room temperature for the first time by our research group.

Temperature is a key parameter for many surface phenomena. Being limited to only one sample temperature for tunnelling microscopy is extremely limiting. Operating at room temperature completely eliminates the possibility for studying many interesting and exciting systems.

Conversely, having control over the sample temperature allows for the possibility
of investigating these phenomena. As described in the literature review chapter, surface phenomena, such as diffusion and phase transitions, are temperature dependent and require sample temperature control to study. For example, Sprunger et al. characterised the growth of Ag on the Cu(100) surface between 150 and 330 K [29]. Below 250 K, the Ag grew in a c(10 × 2) overlayer structure. Above 300K, the Ag formed a substitutional alloy with the Cu(100) surface at coverages below 0.13 ML. At higher coverages, the Ag instead forms small islands of the c(10 × 2) superstructure. Without temperature control, this study would have been impossible.

Investigating the dynamics of diffusing atoms and molecules is difficult when the rate of diffusion is comparable to, or greater than, the scan rate. The rapidly diffusing species can show up as streaks or “fuzziness” in images, as shown in figure 3.1. The amount of thermal energy available to surface atoms and molecules decreases as the temperature is lowered. This results in reduced diffusion rates and, at sufficiently low temperatures, can fix atoms and molecules on the surface. Consequently, they are imaged more clearly and studies into their dynamics become possible.

A commonly used method to achieve variable-temperature operation, involves cooling only the sample and its holder [1, 6, 14, 15, 20]. This leaves the rest of the microscope at, or near, room temperature. Cooling just the sample holder provides a number of benefits over isothermal cooling. Faster temperature changes are possible with a smaller mass being cooled. When the temperature is increased, the sample is less likely to be contaminated by desorbing adatoms or molecules. This is because the surface area that is desorbing contaminants is typically less than that for a comparable isothermal design. Another benefit is that the maximum scan range is not reduced. The microscope tip motion relative to the surface during scanning is controlled with
a piezoelectric tube scanner. Applying a voltage to a piezoelectric material causes it to deform. The magnitude of the deformation is temperature dependent, decreasing with temperature. For example, Gaisch et al. report that piezoelectric sensitivity decreased by a factor of 6.2 between 300 and 10 K [11]. By cooling only the sample, the scan range of the microscope was unaffected.

One of the main challenges faced during the development of the cold stage was incorporating it into an existing microscope. This imposed a number of physical constraints on the cold stage design. The sample holder, transfer mechanism, vibrational damping system, and scanner all had to be accommodated by the new cold stage. Ideally, these components would require few or no modifications. In addition to requiring less work, fewer modifications also meant that there was less downtime during the installation of the cold stage. As the microscope was in use almost every day, any downtime for the system translated into a loss in research productivity.
3.2 Design Goals

Here is a list of the design goals for the cold stage:

- The inclusion and operation of the cooling system should not noticeably increase the level of mechanical noise present at the tip-sample junction.

- It should be possible to operate the microscope at any temperature between the base temperature and room temperature.

- It must be possible to perform atomic resolution scanning over the entire temperature range accessible with the cold stage.

- Any modifications required to the existing system should be limited to the top layer of the stainless steel vibration damping stack.

- A base sample temperature of 100 K should be reached when operating with liquid nitrogen.

- Liquid nitrogen flow at base temperature should be as low as possible. (note: no publications were found from which a valid comparison could be made)

3.3 Description of the Cold Stage

The cold stage was designed to be a non-isothermal low- and variable-temperature sample cooling stage for tunnelling microscopy. Specifically, the cold stage was made for use with the home built microscope designed by Moffat and McLean [21]. Cooling power was provided by a low-noise, open-flow LT-3B-110 Helitran cryostat from
Figure 3.2: Two views of the cold stage mark 1 installed in the vacuum chamber. This version of the cold stage used 0-80 stainless steel screws to mount it to the block massif. The copper braid was made of 0.0762 mm diameter copper wire, which was significantly thicker than in the mark 2 braid. Also, there was no teflon damper.

Advanced Research Systems, Inc\(^1\). This cryostat was chosen because it was designed specifically for ultra high vacuum and low noise applications. It has also been successfully integrated into a microscope by another research group [31].

At this point it is important to note that there were two versions of the cold stage. After the first round of testing, it was recognised that the cold stage, as shown in figure 3.2, was not suitable for microscopy. The mechanical noise levels were unacceptable and imaging with atomic resolution was impossible. The main problem was determined to be the transmission of vibrations down the copper braid. A new braid was constructed and a teflon damper installed to solve this problem. Other areas for improvement were also identified. As a result, the first cold stage was called the “cold stage mark 1”, and a number of changes were made to it. These changes,

\(^1\)http://www.arscryo.com/lt3b.html
along with the new braid, were implemented and then referred to collectively as the “cold stage mark 2”, or just “mark 2”. Both the mark 1 and mark 2 versions of the cold stage will be described here. Two copies of the mark 1 were manufactured at the outset, but both have since been converted into the mark 2 version. The mark 2, shown in figure 3.6, is now used in two of the group’s microscopes.

3.3.1 Materials

Material choices were limited to materials that are ultra high vacuum compatible and have well characterised physical properties at temperatures down to 4 K. The obvious material choice for high thermal conductivity at an acceptable cost was copper. Oxygen-free high-conductivity (OFHC) copper with a purity of 99.99% (4N) has a thermal conductivity of 401 Wm\(^{-1}\)K\(^{-1}\). The thermal conductivity of high purity copper increases by approximately two orders of magnitude as temperature decreases from 100 to 10 K [34]. With the exception of a few small parts and screws, the entire cold stage assembly was made with 4N copper.

At low temperatures, it was important to consider the affect of radiative heat transfer. For two plane parallel surfaces, the net rate of radiative heat transfer in Watts is

\[
\dot{Q} = \sigma A(T_1^4 - T_2^4) \frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2 - \epsilon_1 \epsilon_2},
\]

where \(\epsilon_1\) and \(\epsilon_2\) are the emissivities of the two surfaces at their respective temperatures \(T_1\) and \(T_2\). The area of each surface is \(A\) and \(\sigma\), Stefan’s constant, is given by

\[
\sigma = 5.67 \times 10^{-8}\ \text{Wm}^{-2}\text{K}^{-4}.
\]

From an inspection of the \(T^4\) terms in equation 3.1, one can see that radiative heat transfer becomes significantly more important as the temperature difference increases.
Also, and perhaps more relevant is the fact that lower emissivities reduce the rate of heat transfer. The emissivities for clean surfaces of pure gold and pure copper are both between 0.02 and 0.03 [34]. Gold was preferred for low emissivity applications because copper rapidly oxidises when exposed to atmospheric conditions. The emissivity of oxidised copper is 0.6 [34]. Fortunately, only an extremely thin coating of gold was required and the cost was not prohibitive. As an example of radiative heat transfer, consider a gold plated, 50 mm diameter disc, which is approximately the size of the bottom plate, facing a stainless steel surface. Machined stainless steel has an emissivity of approximately 0.14 \(^2\) [34] and the surface area exposed to the stainless steel is 0.0078 m\(^2\). Using 0.02 as the emissivity for the bottom plate and fixing the temperature of the stainless steel surface at 295 K, the net heat transfer rate was plotted for bottom plate temperatures down to 4 K, as shown in figure 3.3.

Another important material for this project was sapphire. In cases where high thermal conductivity and electrical insulation were both required, sapphire was an ideal choice. While its thermal conductivity was significantly lower than that of copper, it was still much higher than most other ultra high vacuum compatible electrical insulators.

Finally, for a material that is both electrically and thermally insulating one possible choice is polyetheretherketone (PEEK). PEEK is a high performance engineering thermoplastic that, among other desirable attributes, is ultra high vacuum compatible. With a thermal conductivity of only 0.25 Wm\(^{-1}\)K\(^{-1}\), it makes an excellent thermal insulator. PEEK also has excellent mechanical strength in both bending and compression modes. At 213 K, PEEK has a flexural strength of 180 MPa and tensile

\(^2\)http://snap.fnal.gov/crshield/crs-mech/emissivity-eoi.html
yield rating of 105 MPa. Another beneficial characteristic of PEEK is that its physical properties improve as temperature is decreased [7]. PEEK is also machinable allowing it to be used for custom manufactured parts.

3.3.2 The Copper Braid

To provide a thermal connection between the cryostat and the cold stage, a copper braid was employed. The so-called copper “braid” was also not technically a braid, but instead a collection of thin and parallel wires that were intentionally not woven together during its construction. In addition to having excellent thermal properties, high purity copper is very soft. Such copper braids are known transmit vibrations poorly and have been used before in low temperature microscopes [1, 6, 14].
A number of precautions were taken to ensure that vibrational noise transmitted by the braid would be small. First, high purity copper wire with a small diameter was used to construct the braid. The original braid was 7 cm in length. It was assembled from 1200 copper wires, each with 4N purity and a diameter of 76.2 µm. The resulting cross-sectional area of the braid was 5.47 mm$^2$. Using silver solder, the braid was fixed to copper blocks, each measuring approximately 30×15×3.5 mm$^3$. These small blocks had holes in them and they were in turn screwed to the cryostat and the bottom plate of the cold stage. This copper braid can be seen in figure 3.2 connecting the cryostat tip to the bottom plate of the cold stage mark 1.

Unfortunately, the mechanical coupling between the cold stage and the cryostat was too strong with this braid. Consequently, it was not possible to image the surface with atomic resolution. A second braid was made from 9000 wires of copper with the same purity (4N), each with a diameter of 25.4 µm. The new braid has the same overall length as the first braid, but a reduced cross-sectional area of 4.56 mm$^2$. With the first braid, it was observed that the solder had wicked up the braid, this stiffened the braid. Instead of soldering, the second braid was clamped between the copper blocks at either end. This ensured that the braid remained soft and allowed for future changes to be made to the braid more easily.

### 3.3.3 Key Design Features

In addition to sample cooling, the cold stage has a number of other important and useful features. These new features mark it as a significant improvement over the existing sample stage in terms of both functionality and ease of use.

One particularly significant addition was a mechanism for clamping the sample
Figure 3.4: A simplified “section view” through the centre of the cold stage (A), the plunger (B), the block massif (C), the sample holder (D), and the screws connecting the cold stage to the block massif. For the mark 1, the screws (E) were 0-80 stainless steel screws. These were changed to slightly large diameter PEEK screws for the mark 2 to reduce the thermal connection between the cold stage and the block massif.

holder against the sample stage. The sample holder is used to facilitate the transfer of the sample to different parts of the system. For scanning, the sample holder is moved into the cold stage, where it is now held by a clamp. This makes transferring the sample holder on to and off of the cold stage a much more robust and reliable operation. The clamping mechanism, called the “plunger” because of its shape and motion, is shown in figure 3.4. Previously, extensive use of a wobble stick was required during transfers. The wobble stick was used to fix the sample holder in place during transfers or to push it off of the sample stage. The transfer fork grips the sample holder more tightly than the cold stage does causing the sample holder to move with the fork. With the plunger, one simply brings the sample holder onto the cold stage, then engages the plunger using a 1” linear push-pull and retracts the fork. A spring applies an upward force through the plunger that fixes the sample holder in place,
pressing it firmly against the molybdenum tables that the beetle walks on. This force also provides a good thermal contact between the cold stage and the sample holder.

To assist in sample transfer, the leading edge of the cold stage presented to the sample was offset as shown in figure 3.5. This compensates for small vertical misalignments between the cold stage and the transfer arm. Misalignments are very difficult and time consuming to completely eliminate. The offset leading edge allows one side of the sample holder to enter the cold stage before the other. At this point, the transfer arm can be rotated to bring the other side of the sample holder into alignment with the cold stage, if it wasn’t already. Previously, any such vertical misalignments would require adjustments to the entire transfer system. This is, therefore, a significant improvement.

The tables that the beetle walks on were removed from the sample holder and made part of the cold stage. As part of the sample holder, the tables would routinely be covered in thin films and were very close to the sample when it was heated for cleaning. Over time this exposure would lead to a dirty walking surface for the beetle, which can cause problems when making coarse movements with the beetle. By moving the table to the cold stage, this problem was eliminated.

As part of the mark 2 upgrade to the cold stage, a teflon clamp was added to fix the braid half-way along its length. This has been shown to significantly reduce vibrational noise transmitted through the braid [6, 14]. The teflon clamp was fixed rigidly to the block massif. Teflon has a low thermal conductivity of only 0.27 Wm\(^{-1}\)K\(^{-1}\) \(^3\), so heat transfer through the teflon clamp was expected to be insignificant. The contact areas between the copper braid and the teflon clamp, and between the teflon clamp and the block massif were minimised to further reduce any heat transfer between the

\(^3\)http://www.yutopian.com/Yuan/prop/Teflon.html
Figure 3.5: A top view of at the cold stage (F), the transfer fork (C), and the sample puck (E). Note, in particular, that one edge of the dock (A) on the cold stage protrudes farther than the other (B). The sample (D) is mounted on the sample puck (E). The braid [not shown] is fixed to the bottom plate of the cold stage at the tab (G). For clarity, the tables on the top of the cold stage have been removed from this diagram.
two masses.

One of the interesting problems was the need to have a strong thermal link between the cryostat and the sample, but to also have them electrically isolated. To perform tunnelling microscopy, a bias needs to be applied between the microscope tip and the sample. To reduce electrical noise, this circuit needs to be electrically isolated from everything else; including the chamber, which is grounded. The cryostat was in electrical contact with the chamber, so there needed to be an electrical break at some point between the cryostat and the sample holder. The contacts at either end of the braid were going to be small, so those locations were not ideal for the break. Also, the mark 1 version of the cold stage was fixed to the block beneath with 0-80 stainless steel screws. Taking all of this into consideration, it was decided to design the cold stage to have separate top and bottom sections. The top half was in electrical and mechanical contact with the sample holder, but was electrically isolated from the bottom half. A 0.5 mm sapphire plate was machined to sit between the top and bottom halves of the cold stage. As discussed earlier, sapphire is both a good thermal conductor and a good electrical insulator. This arrangement allowed for a large contact surface area between the sapphire and the copper pieces.

A prominent difference between the mark 1 and mark 2 versions of the cold stage, was the use of PEEK as a replacement for many of the stainless steel parts. In particular, the 0-80 pushing and pulling screws that fixed the cold stage to the block massif where replaced by PEEK. The size of the “pulling” screws was increased to 2-56, while the number of screws was reduced from 6 to 3. The 6 “pushing” screws were replaced by 3 PEEK pins of the same diameter. This change reduced the thermal connection between the cold stage and the block massif by more than an order of
Figure 3.6: The cold stage mark 2 (A) installed in the STM chamber. For the cold stage mark 2, custom made 2-56 PEEK screws and pins are used to mount it to the block massif. The copper braid (E) is made of finer [25.4 µm diameter] wire and there is a teflon damper (C). To maintain the softness of the braid, it was clamped to the copper blocks (B), instead of soldered, like the mark 1 braid. The cryostat tip (D) has a threaded hole in end to which one end of the braid is bolted into.
magnitude. The pins on the end of the plunger were also changed to PEEK. There were some problems with the stainless steel pins touching the bottom plate, shorting the sample holder to the bottom plate. The use of PEEK pins resolved this issue.

With the mark 2, the cold stage was fixed to the block massif by 3 2-56 screws and 3 0-80 sized pins, all made of PEEK. The flexural strength of the PEEK parts was checked against the maximum force that could be applied by the magnetic transfer arm. It was determined that the PEEK replacements were sufficiently strong and would not break during regular usage.

All of the copper parts on the entire cold stage were given a thin gold plating. As mentioned earlier, the gold plating ensures that the surface retains a low emissivity by not oxidising when exposed to atmospheric conditions. Without the gold plating, the copper would tarnish and oxidise over time, significantly increasing its emissivity.

### 3.4 Thermal Modelling

During the design process it is often informative to make a physical model. A model can show the effect that varying important parameters has on the performance. For the cold stage, basic thermal modelling was performed to determine what would most influence the base temperature of the sample and the time it would take to cool the sample to base temperature.

In order to determine the cool down time and minimum temperature of the sample, a thermal model of the cryostat and cold stage assembly was created. The high thermal conductivity of copper leads naturally to using a “lumped” heat capacity analysis. The main assumption made was that there were no significant thermal gradients within each individual part of the assembly. This analysis method can
be applied to systems where the materials present have high thermal conductivities and high contact resistances. Figure 3.7, shows the separate parts of the cold stage included in the model. The most important thermal contacts were expected to be between the copper braid and the bottom plate, and also between the top plate and the sample holder. These contacts, with low surface area and/or low contact pressure, tend not to conduct heat well. This is especially true in vacuum, where there is no medium filling the microscopic gaps. On the other hand, connections with high contact pressure and surface area, such as those between the top plate, the sapphire, and the bottom plate, were expected to conduct heat well.

### 3.4.1 Convective Heat Transfer

As already discussed, radiative heat transfer is important, and as such, it was accounted for in the model. Equation 3.1 was used to estimate the net radiative heat
transfer for each component of the cold stage. On the other hand, convective heat transfer was determined to be insignificant. At low pressures, heat transfer between two approximately parallel surfaces via conduction through a gas is given by

\[ \dot{Q} \approx \text{constant} \times a \times P(T_2 - T_1), \]  

(3.3)

where: \( a \) is the accommodation coefficient, \( P \) is the pressure in Pa, and \( T_1 \) and \( T_2 \) are the temperatures of the two surfaces [34]. The resulting heat transfer, \( \dot{Q} \), is given in Wm\(^{-2}\). The value of the constant ranges from 1.2 to 4.4 depending on the gas, while the accommodation coefficient ranges from \( 0.3 < a < 1.0 \). In our system, the pressure is typically lower than \( 10^{-8} \) Pa, so even with a temperature difference as large as 190 K, \( \dot{Q} \) was still only on the order of \( 10^{-5} \) Wm\(^{-2}\). Then, using the surface area from the radiative example earlier, 0.0078 m\(^2\), the heat transfer value went down two orders of magnitude, and was insignificant compared to even the radiative heat transfer.

### 3.4.2 Thermal Contact Resistance

For this model, the contact resistance between the parts was one of the largest factors in determining the cool down time of the sample holder. Estimating contact resistance values was very difficult, as they rely on a number of properties that were hard to determine. Thermal contact resistance values given in tables were used initially, but as each contact will have different particulars, they were mostly useful only as a rough estimate. The first estimates for contact resistances were based on known resistances for metallic interfaces under vacuum [16, 32], and are shown in table 3.1.

Later, an article was found that reported a method used in microchips to calculate interface resistances at low contact pressures [37]. The calculation presented includes such factors as surface roughness, microhardness, thermal conductively of
both interface materials, and gap substance (if any). The contact conductance is given by,

\[ h_c = 1.25k_s \frac{m}{\sigma} \left( \frac{P}{H_c} \right)^{0.95}, \]

where: \( k_s \) is the harmonic mean thermal conductivity of the interface materials, \( m \) is the effective mean absolute asperity slope of the interface, \( \sigma \) is the RMS surface roughness, \( P \) is the contact pressure, and \( H_c \) is the surface microhardness of the softer of the two materials. Since the surface asperity slope is not typically available, it can be estimated as,

\[ m = 0.125(\sigma \times 10^6)^{0.95}, \]

when the surface roughness is in the range of 0.216 \( \mu m < s < 9.6 \mu m \). Using this method, new thermal contact resistances were determined. These values depend on the thermal conductivity of the interface materials, which varies with temperature, so they were calculated with each step of the program. For comparison, the thermal contact resistance values for the interfaces, calculated at 295 K using equation 3.4 are shown in table 3.2.

With thermal resistance values available, the heat flux between the parts of the model were calculated using the formula,

\[ \dot{Q} = \frac{\Delta T}{R/A}, \]
Table 3.2: Sample contact resistance values at 295 K calculated using equation 3.4.

<table>
<thead>
<tr>
<th>Interface</th>
<th>Contact Resistance (cm$^2$KW$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Copper Braid Block - Bottom Plate</td>
<td>18</td>
</tr>
<tr>
<td>Bottom Plate - Sapphire Plate</td>
<td>1.8</td>
</tr>
<tr>
<td>Sapphire Plate - Top Plate</td>
<td>0.7</td>
</tr>
<tr>
<td>Top Plate - Sample Holder</td>
<td>37</td>
</tr>
</tbody>
</table>

where $\Delta T$ is the temperature difference between the surfaces in K, $R$ is the thermal contact resistance in cm$^2$KW$^{-1}$, and $A$ is the contact area of the interface in cm$^2$.

### 3.4.3 The Model

The thermal model of the system was obtained by calculating the net heat transfer from both conduction and radiation between each part of the system. The cold stage was separated into parts by dividing it at each thermal interface, as shown previously in figure 3.7.

With an estimate for the total heat flux, the temperature change in a single part can be calculated using the following formula,

$$\Delta T = \frac{\Sigma Q}{Cm} \Delta t,$$

where $m$ is the mass, $\Sigma Q$ is the sum of all of the heat fluxes acting on the part, $C$ is the specific heat capacity of the material, and $\Delta t$ is the time over which the heat transfer occurs. There is no x-dependance because the “lumped” heat capacity model assumes no temperature gradients within individual parts.

A Matlab program was created to perform this calculation iteratively, determining the heat flow to and from each part of the model. The temperature of the cryostat tip was assumed to be at the temperature of liquid nitrogen, 77 K. The program was run until the temperature of each part had equilibrated.
Figure 3.8: Cooling curves produced by the computer model using the contact resistance values from table 3.1. In 1500 minutes, the sample holder reaches a base temperature of 84 K. Note that the curves for the bottom plate, the sapphire plate, and the top plate all lie together and are indistinguishable.

3.4.4 Modelling Results

Using the program, one cooling curve was produced for each set of thermal resistance values. In both cases, the data for the bottom, sapphire, and top plates all overlap and as such are indistinguishable on the plots. Figure 3.8 corresponds to the data generated using the thermal contact resistance values from table 3.1, while 3.9 corresponds to the data generated using the thermal contact resistance values calculated using equation 3.4.
Figure 3.9: Cooling curves produced by the computer model using equation 3.4 to calculate the contact resistance values. In just 43 minutes, the sample holder reaches a base temperature of 78 K. Again, the curves for the bottom plate, the sapphire plate, and the top plate all overlap.
Table 3.3 summarises the results from both data sets generated by the thermal model of the cold stage. As a useful performance metric, the time constant was determined for each data set from an exponential decay fit to the data.

Table 3.3: The results of thermal modelling.

<table>
<thead>
<tr>
<th>Resistance values</th>
<th>Base temp. (K)</th>
<th>Cooling time (min)</th>
<th>Time const. (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Table</td>
<td>84</td>
<td>1500</td>
<td>401.7±0.2</td>
</tr>
<tr>
<td>Equation</td>
<td>78</td>
<td>43</td>
<td>11.36±0.04</td>
</tr>
</tbody>
</table>

These results clearly demonstrated the importance that thermal contact resistance will have on the performance of the cold stage. In both situations, the bottom, sapphire, and top plates were always at the same temperature. Additionally, both cases also showed temperature differentials between the braid and the bottom plate, as well as, between the top plate and the sample holder. Based on these results, it was recognised that these two interfaces would be very important in determining the cooling performance of the cold stage. Consequently, significant effort was put into lowering the thermal contact resistances at these interfaces. Gold foil was put in the braid to bottom plate interface. Gold is very malleable and soft, and filled any microscopic gaps. All contact surfaces were also carefully flattened and polished to maximise the contact area. For the top plate to sample holder interface, the contact pressure was made as high as possible by selecting a strong spring for the plunger.

### 3.5 Testing

The cryostat that provides the cooling power for the cold stage can operate using both liquid helium and liquid nitrogen. Though liquid helium produces a lower base
temperature, the cost of operating with liquid helium is prohibitive [\(\sim \$10 \text{ L}^{-1}\)]. Liquid nitrogen is a much more cost-effective alternative, at only \(\sim \$0.60 \text{ L}^{-1}\) \(^4\). With this in mind, liquid nitrogen is expected to be used the vast majority of the time, and so testing was performed with liquid nitrogen.

3.5.1 Goals

Testing was carried out on the cold stage to determine its cooling performance and to ensure that cooling did not have a detrimental impart on image acquisition.

The cold stage was tested with liquid nitrogen with the following objectives in mind.

- The base temperature of the cold stage and sample should be established.
- The cooling curve for the cold stage and the time constant for cooling should be determined.
- The cryogen flow (consumption) rate at base temperature should be measured.
- It should be verified if any temperature gradients exist across the cold stage. This would identify areas for future improvement.
- Atomic resolution STM images should be collected with the sample at base temperature.

\(^4\)The prices for liquid helium and liquid nitrogen were obtained from the physics department stores at the time this thesis was written.
3.5.2 Experimental Setup

Initial testing of the mark 1 cold stage was carried out in an ultra high vacuum chamber from the decommissioned KRIPES system. The geometry of the relevant ports on this chamber was very similar to that of the STM chamber. Most importantly, relative to where the cold stage sits, the position and angle of the port that the cryostat was mounted to is the same in both chambers.

On this chamber, there were enough ports for all of the UHV thermocouple feedthroughs. K-type thermocouples were attached to each section of the cold stage: on either end of the braid, the bottom plate, the top plate, and, when available, the sample.

3.5.3 Liquid Nitrogen Testing

Initial testing found that the original copper braid coupled the external environment too strongly to the cold stage. With the resulting mechanical noise, atomic resolution imaging with the STM was impossible. As described earlier, a number of changes were made to the braid to reduce the noise to acceptable levels. The cold stage mark 1 has subsequently been upgraded into the mark 2 and tested. With the mark 2 version, Andrew Mark has successful taken atomic resolution images at the base sample temperature. Only cooling results from the cold stage mark 2 will be presented here, as these are the relevant results. Thermally, both versions of the cold stage performed very similarly.

As seen in figure 3.10, the cold stage reaches a base temperature of 109 K in approximately 300 minutes (5 h). Previous tests have shown that the maximum temperature difference between the sample temperature and the cold stage temperature
CHAPTER 3. VARIABLE-TEMPERATURE STM SAMPLE STAGE

Figure 3.10: Cooling curves from the cold stage mark 2 operating with liquid nitrogen. The cold stage reached a base temperature of 109 K, which corresponds to a sample temperature of approximately 114 K.
is less than 5 K. This would give a maximum sample temperature of 114 K. During normal operation, measuring the sample temperature directly is impossible, so it must be inferred from the measured temperature of the cold stage. About 10 minutes after the cryogen transfer line was cooled, the cryostat was at its base temperature, and the cryogen flow rate reduced to less than 0.16 Lh$^{-1}$ for the remainder of the cooling. With a 60 L dewar, this would allow experiments to last for over 15 days.

The cooling curve in figure 3.10 exhibited an exponential decay. To determine the time constant for the cold stage, the “cool down” section of the cooling curve was fit with a decaying exponential of the form,

$$T = A \exp(-t/t_0) + T_0,$$

where: $T$ is the temperature in K, $A$ is a fitting constant, $T_0$ is the initial temperature, $t$ is the time in minutes, and $t_0$ is the time constant for cooling. This resulted in an excellent fit with $\chi^2 = 0.72$, as shown in figure 3.11. From the fit, the time constant, $t_0$, was found to be 61.17±0.03 minutes.

To determine the impact that cooling has on the mechanical noise at the tip-sample junction, a Si(111) sample was cooled and imaged on the mark 2. Significant thermal drift was experienced while the temperature of the sample changed. At 159 K, the sample temperature was changing at a rate of approximately −0.1 K/min and the resulting drift can be seen in figure 3.12.

After cooling for 5 hours, the cold stage was at 109 K and thermal drift had decreased significantly. A image of the surface at 114 K can be seen in figure 3.13. The individual adatoms of the Si(111)-7×7 reconstruction were clearly distinguishable from one another. The ability to resolved the individual silicon adatoms demonstrated that the noise levels were low enough for atomic resolution imaging.
Figure 3.11: Equation 3.8 was fit to the cooling data in figure 3.10. From the fit, a time constant was found to be $t_0 = 61.17 \pm 0.03$ minutes.
Figure 3.12: Two images of a Si(111)-7×7 taken using the cold stage mark 2 at 159 K. 3.12a was taken with the fast-scan direction in the horizontal direction and 3.12b was taken with the fast-scan direction in the vertical direction. These images were acquired by Andrew Mark while the sample was being cooled at $\sim -0.1$ K/min. Very high levels of drift can be seen in both images. In (a), the brighter features should be triangular islands, but appear flattened vertically because of drift. In (b), the long, bright streaks are more triangular islands that appear stretched out because of the drift. These images indicate that the differential thermal expansion is too high during temperature changes to acquire useful images.
This surface was prepared as per the typical procedure for creating Si(111)-7×7. It was then exposed to approximately half of a bi-layer of germanium and finally annealed for 15 minutes. In an attempt to reproduce the results of Stipe et al. [30], scanning was performed at high voltage. This was expected to produce displaced adatoms on the surface. In addition to the expected 7×7 pattern, a number of bright features were also observed. One image is shown in figure 3.13 and a few examples of the displaced adatoms have been circled. Just before this image was taken, a pulse at 3.4 V at 10 nA for 100 s was applied to the surface. Prior to the pulse, only the 7×7 reconstruction was observed, with no displaced adatoms. This clearly demonstrates that it is now possible to study processes that can’t be observed at room temperature. At room temperature, the adatoms would recombine with the adjacent adatom vacancies before they could be observed with the microscope.

3.5.4 Summary

The cold stage successfully extends the operating temperature of the scanning tunnelling microscopy systems it was integrated into below room temperature. Though the desired base sample temperature of 100 K was not attained, the cold stage’s base sample temperature of 114 K still comparable to a similar instrument’s base sample temperature of 116 K [22]. With the improvements made to the second version of the cold stage, atomic resolution scanning was performed at low temperatures. The other improvements that were part of the cold stage design have reportedly been very helpful, making the system easier to use and more robust. The plunger and offset leading edge both provide easier and more robust sample transfer. Since the tables have been moved to the cold stage, there have been no reported problems with the
Figure 3.13: An image of a Si(111)-7×7 surface at a temperature of \( \sim 114 \) K. White circles indicate features similar to those observed by Stipe et al. [30] and are likely either displaced Si or Ge atoms. Stipe et al. showed that these displaced atoms are metastable up to 155 K. At higher temperatures, they return to their original position in the surface. Inset: image of the surface before the 3.4 V at 10 nA pulse was applied for 100 s. Scanning parameters: +1.5 V at 146 pA. Images taken by Andrew Mark.
beetle walking.

At this time, only one experiment has been performed that has made use of the low-temperature capabilities of the cold stage. This experiment clearly demonstrated the ability of the cold stage to provide both sample cooling and the necessary low noise environment necessary for tunnelling microscopy.

3.6 Recommendations

Two obvious improvements for the cold stage would be to lower the base sample temperature and to increase the cooling rate of the sample. In Michael Schumack’s design [22], when using liquid nitrogen, samples are cooled down to 116 K approximately 60 minutes after the transfer line has cooled. While we have not achieved the same cooling rate, our base sample temperature is very similar. With the mark 2 braid, vibrational noise is no longer appears to be an issue.

Again, it is a matter of how much weaker can the mechanical connection be made before it will mechanically fail. One obvious change would be to decrease the diameter of the 4 PEEK poles on the plunger. PEEK is quite strong against compressive forces, so the diameter of these poles can be reduced. Weakening any thermal connections between the cold stage and the block massif would also have a positive improvement on cooling performance. With the mark 2, the cold stage is fixed to the block massif by 3 2-56 screws and 3 0-80 sized pins, all made of PEEK. This was determined to be sufficiently strong, so as not to break during regular usage.

It is possible that more wires could be added to the braid, without having a significant negative impact on imaging. Temperature measurements at the teflon damper indicate that it is not being cooled much below room temperature, but it
still is a source of heat gain to the system. Insulating it more from the block and the braid would also have a positive influence on cooling. Also, in other designs with damping on a copper braid \[6, 22\], the damper is fixed to a secondary mass, separate from the mass the sample is fixed to. If a thicker braid was used, it would provide a stronger thermal connection between the cryostat and the cold stage. This would allow faster temperature changes and should also reduce the base temperature of the cold stage.

The cold stage also has the possibility to operate as a variable-temperature stage. Time constraints on the instrument prevented this from being tested. There are two possible methods by which this could be accomplished. First, the temperature of the continuous flow cryostat used with the cold stage can be controlled. By changing the temperature of the cryostat, the temperature of the cold stage would also indirectly be controlled. This would allow variable-temperature operation with no modifications to any of the equipment, though temperature change is expected to be rather slow. Alternatively, or perhaps in combination with the first method, a heater diode could be fixed to the cold stage. The diode would provide counter-heating to the cold stage. By directly heating the cold stage, temperature changes would be rapid.

If the requirement that the cooling stage must work with existing technology were lifted, then more radical changes could be made that would likely result in significantly increased cooling performance. Perhaps the most restricting was the requirement to accommodate the existing sample holder. For example, if the sample holder from the new microscope being developed were used instead, the entire cold stage could be scaled down with it. The location of the electrical break could also be moved to either end of the braid. Only a thin sapphire washer would then be needed, instead of a
custom machined sapphire part. There would no longer be separate top and bottom plates, again reducing the size and mass of the cold stage.
Chapter 4

Isothermal Scanning Tunnelling Microscopy Cold Cans

4.1 Introduction

The benefits of operating a scanning tunnelling microscope at low temperature have already been discussed in a previous chapter. This chapter will focus on the radiation shields and cooling system, hereafter collectively referred to as the “cold cans”, for the new scanning tunnelling microscope head currently being designed by Drevniok and McLean. The cold cans were designed to operate isothermally; that is, with the microscope head and sample at the same temperature. They also provide an ultrahigh vacuum compatible and thermally shielded environment for the microscope to operate in. For rapid cooling and during tip or sample exchange, the cold cans act as a dock for the microscope. A number of other cooling systems \[11, 20, 26, 31\] were investigated and used as guides for the design of the cold cans. In particular, the design presented by Stipe et al. was drawn upon heavily \[31\].
Cooling systems for scanning tunnelling microscopes can be a challenge to design and implement. The requirement for a strong thermal path between the cryostat and the microscope competes with the need to minimise the transmission of vibrations to the microscope. Open cycle cryostats couple the chamber to an external Dewar introducing a new source of vibrational noise to the system. This drawback is offset by the ability to control the sample temperature from room temperature, or higher with active heating, down to the base temperature of the cryostat. Although bath cryostats do not have the same external noise coupling, they provide only limited temperature control. The base temperature is determined by the cryogen, the strength of the thermal connection, and the heat load on the mass being cooled. To ensure a low base temperature for the sample and microscope, all external connections need to be heat sunk before reaching the microscope.

4.2 Design Goals And Requirements

Here is a list of the design goals for the cold cans.

- The base sample temperature should be less than or equal to 8 K.
- It should be possible to control the temperature between base temperature and room temperature with a temperature fluctuation of less than 0.1 K, relative to the set point.
- It should be possible to image with the microscope at any temperature accessible with the cryostat.
- The design should easily accommodate future additions; such as light collection apparatus and gas dosing access.
• The cryogen consumption rate should be as low as practically possible for an open-flow cryostat design.

• The design should allow in-vacuum physical access to the microscope for sample and tip exchange that does not compromise the base temperature or cryogen consumption rate.

4.3 Description of the Cold Cans

4.3.1 Design

The cold cans comprised of two nested copper radiation shields. These cans were connected only through insulated parts of the cryostat head, and as such, do not have a strong thermal connection. The inner can was connected to the tip of the cryostat head which was cooled directly by the cryogen. The outer can was cooled by the exhaust cryogen gas and provides a second radiation shield. This reduced the cryogen consumption rate, and also resulted in a lower base temperature for the inner can.

The top, back, and bottom sides of each can were made from a solid piece of copper. Replaceable panels covered the remaining 3 sides. The solid copper piece ensures excellent thermal conductivity between the cryostat tip and microscope. The replaceable panels accommodate future modifications, so that changes, at most, require a new panel to be machined. The panels for the outer can were 1 mm thick to reduce the mass being cooled.

Vertically sliding doors on the front panel granted access to the microscope for tip exchange, sample exchange, and visual inspection of the tip-sample junction.
Figure 4.1: Four images of the cold cans during assembly. (A) The side panels of the outer can and the return cryogen flow shield of the cryostat have been connected to the back of the outer can. (B) The back of the inner can, with its side panels connected, has been mounted to the tip of the cryostat. (C) The front panel of the inner can was attached to the inner can. (D) Finally, the front panel of the outer can was attached to the outer can.
The doors were machined from aluminium to ensure that no cold welds would form between the door and the can.

A sapphire “pin panel” was mounted to the bottom on the inner can. All wires running to the microscope were thermally anchored to the sapphire pin panel. Thermally anchoring the wires reduces the heat load on the microscope [31].

4.4 Testing

Testing was carried out on the cold cans to determine their cooling and temperature control performance.

4.4.1 Goals

For both liquid helium and liquid nitrogen the cold cans were tested to obtain the following list of parameters and objectives:

- Find the base temperatures for the inner and outer cans.
- Determine the time it takes to cool the cold cans from room temperature to base temperature.
- Measure the cryogen flow (consumption) rate at base temperature.
- Demonstrate and quantify the precision, stability, and range of temperature control available.
- Check to see if any temperature gradients exist across each can.
4.4.2 Testing Setup

The cold cans were mounted in the lab’s “test” chamber. This chamber was pumped by a 60 Ls\(^{-1}\) turbo molecular pump mounted on a 4-1/2” CF flange. A pressure of 1.5\(\times\)10\(^{-7}\) Torr was achieved in the chamber, and this was sufficient for the operation of the cold cans. Upon cooling, the cold cans provided additional pumping, reducing the pressure to 8\(\times\)10\(^{-9}\) Torr. A suitable 6” CF flange port was available to mount the cryostat and cold can assembly. Two 2-3/4” CF flange ports were conveniently located near to the cold cans for thermocouple feedthroughs.

K-type thermocouples were mounted in various arrangements at key locations on the cold cans and passed through an ultra high vacuum thermocouple feed through. Initially, only three thermocouple feedthroughs were available, but for the final liquid
helium run extra feedthroughs were purchased to allow ten thermocouples to be used. The cryostat head came equipped with two silicon diode temperature sensors. One was permanently attached to the tip of the cryostat while the other was free to be placed at any desired location within a few inches of the cryostat tip. The reading from the silicon diodes were found to be more reliable than the k-type thermocouples in general, and particularly below 50 K. As such, the second Si diode was placed in locations of high importance like the back of the inner can close to where the microscope head would dock.

Outside of the chamber, the thermocouple leads were attached to a National Instruments TBX-68T connector block. This block provides a built-in cold-junction compensation sensor to correct for the voltage generated at the junctions between the thermocouple leads and screw terminals on the block. A shielded cable connects the block to a NI PXI-4351 card that is sitting in a PXI chassis which is interfaced to a Windows-based computer running Labview 7.1.

A custom Labview program collected the thermocouple voltages, the voltage from the cold junction thermistor, and the temperatures from the Si diodes as read by the Temperature Controller. The program applied the cold-junction compensation voltage to each thermocouple voltage, and then converted the voltages into temperature using a formula and coefficients provided by the National Institute of Standards and Technology [NIST]$^1$. All data were time stamped and saved to a tab-delimited text file for later analysis. Temperature data were plotted in real-time by the program.

The circuit for gas and cryogen flow is shown in figure 4.2. A specialised cryostat system consisting of a vacuum shielded transfer line, the cryostat head, a Dewar adaptor, the flow meters, the temperature controller, and the required plumbing was

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$^1$http://srdata.nist.gov/its90/type_k/kcoefficients_inverse.html
purchased from ARS\textsuperscript{2}. This particular cryostat was well suited for use with a scanning tunnelling microscope because it generated very little mechanical noise, was UHV compatible, and provided access to a wide range of temperatures. Precision flow control was an important factor in temperature control and minimising the consumption rate of the cryogen. The high cost of liquid helium [approximately $10 \text{ L}^{-1}$ from Physics Stores] required that any flow beyond the minimum necessary to maintain the target temperature be reduced as much as possible. The cryogen flow was controlled in three ways: by changing the Dewar pressure, an adjustable needle valve in the tip of the cryostat, and a flow meter that monitored and limited the flow through the head and shield of the cryostat. Dewar pressure was the most crude method of flow control. During the first stage of cooling, the Dewar was pressurised to 5.0 psi. After the tip has reached its minimum temperature, the Dewar pressure was reduced to 2.5 psi. Since the outer can was cooled by the exhaust flow, reducing the flow resulted in a longer cool down time for the outer can. At the expense of efficiency the most rapid cool down was achieved by maintaining the maximum cryogen flow at a Dewar pressure of 5.0 psi. After base temperature was attained, the goal was to find the minimum flow rate that provided a stable temperature. Using the needle valve, the flow was reduced as much as possible until the temperature became unstable. Final fine adjustments were made by limiting the flow through the flow meters.

Away from base temperature, control was best obtained by using the heater in the cryostat tip. The temperature controller applied power to the heater based on a PID (proportional integral derivative) feedback control loop. It automatically adjusted the parameters to reach the programmed temperature quickly and remain stable at that

\textsuperscript{2}http://www.arscryo.com/
temperature. Using the heater in conjunction with the controller, it was possible to reach any temperature between room temperature and the base temperature. Away from base temperature it was desirable to reduce cryogen flow to reduce the cost of operation.

When temperatures below 78 K are desired, liquid helium was used. The cryostat and cold cans were pre-cooled with liquid nitrogen to reduce cost. Switching over from liquid nitrogen to liquid helium was a simple task and takes approximately 10 minutes.

4.4.3 Results

The most important location for temperature measurements was the back on the inner can because this was where the microscope will make contact. It was assumed that the microscope will reach the same temperature as the back surface of the inner can. This assumption was based on the idea that the mass of the microscope is very small relative to the inner can and the thermal connection while docked should be good. Throughout all of the trials, it was found that there were no measurable thermal gradient across either the inner or the outer can. In addition, the measured temperature of the inner can was found to be the same as the tip of the cryostat within the measurement uncertainty. These observations allowed us to use the Si diodes that are connected to the cryostat tip to measure the temperatures of the inner and outer cans.

The first test of the cold cans was done with liquid nitrogen. After the cryogen Dewar was pressurised, it took approximately 50 minutes for the cryostat tip to reach a base temperature of 78.4 K. During the first 10 to 15 minutes only negligible cooling
Figure 4.3: This plot shows liquid nitrogen cooling data for the cold cans. The first section shows representative cool down curves for the inner and outer cans. The remainder of the plot demonstrates the temperature control available over the cold cans. Details for each plateau are presented in table 4.1.
is observed as the cryogen was boiling off in the transfer line before it reaches the tip. The outer can takes significantly longer to obtain its base temperature and was not as cold, reaching a temperature of 82.0 K. This behavior was expected since the outer can is cooled by the exhaust cryogen gas from the tip, and not directly by the liquid. 

With no active temperature control, a base temperature of 78.4 K was maintained for 2 hours, but with as much drift as ± 0.4 K. To maintain this temperature, 0.19 Lh$^{-1}$ of liquid nitrogen was consumed. Using the temperature controller and reducing the cryogen flow, the temperature range accessible and the temperature stability were examined. With the temperature controller, stability was greatly improved and temperatures could be held to within ± 0.1 K. Table 4.1 summarises the results of liquid nitrogen cooling.

Table 4.1: Liquid nitrogen cooling results summary.

<table>
<thead>
<tr>
<th>Setpoint (K)</th>
<th>none</th>
<th>80</th>
<th>100</th>
<th>150</th>
<th>200</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average (K)</td>
<td>78.44</td>
<td>80.00</td>
<td>100.00</td>
<td>150.00</td>
<td>200.00</td>
</tr>
<tr>
<td>Min (K)</td>
<td>78.11</td>
<td>79.95</td>
<td>99.88</td>
<td>149.98</td>
<td>199.99</td>
</tr>
<tr>
<td>Max (K)</td>
<td>78.79</td>
<td>80.11</td>
<td>100.09</td>
<td>150.05</td>
<td>200.04</td>
</tr>
<tr>
<td>Standard Deviation (K)</td>
<td>0.12</td>
<td>0.02</td>
<td>0.01</td>
<td>0.01</td>
<td>0.00</td>
</tr>
<tr>
<td>Time at Temperature (m)</td>
<td>120.5</td>
<td>23.5</td>
<td>142.5</td>
<td>37.6</td>
<td>60.8</td>
</tr>
<tr>
<td>Flow Rate (LL/Hr)</td>
<td>0.19</td>
<td>0.15</td>
<td>0.15</td>
<td>0.07</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

As mentioned, the liquid helium run was pre-cooled with liquid nitrogen. During the switch over the inner can temperature had drifted up to approximately 94 K. It took 7.5 minutes for the temperature to reach a base temperature of 5.015 K from 94 K. A temperature of 5.015 K was maintained for 20 minutes with a variation of less than ± 0.023 K. Operating at this temperature required a liquid helium flow rate of 2.65 Lh$^{-1}$, which was higher than the rate of 1.5 Lh$^{-1}$ reported in the manual.
Using the temperature controller, the temperature was raised to 6.00 K. This temperature can be maintained with a reduction in liquid helium flow and requires 1.9 Lh\(^{-1}\). Though the temperature had not fully stabilised and was only maintained for 8 minutes, it did not vary by more than \(\pm 0.10\) K. At 10 K, the flow was reduced again and required only 1.3 Lh\(^{-1}\). Over a period of 4 minutes the temperature was stable to within \(\pm 0.03\) K. Finally, to demonstrate temperature control in the range between 5 and 78 K, the temperature controller was set to 50.00 K and held there for 12 minutes. At 50.00K, only 0.3 Lh\(^{-1}\) was necessary to hold the temperature stable to better than \(\pm 0.02\) K. Table 4.2 summarises the results from the liquid helium testing.

<table>
<thead>
<tr>
<th>Setpoint (K)</th>
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<th>10</th>
<th>50</th>
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<tbody>
<tr>
<td>Average (K)</td>
<td>5.015</td>
<td>6.00</td>
<td>10.00</td>
<td>50.02</td>
</tr>
<tr>
<td>Min (K)</td>
<td>5.010</td>
<td>5.95</td>
<td>9.97</td>
<td>50.00</td>
</tr>
<tr>
<td>Max (K)</td>
<td>5.038</td>
<td>6.09</td>
<td>10.02</td>
<td>50.02</td>
</tr>
<tr>
<td>Standard Deviation (K)</td>
<td>0.004</td>
<td>0.03</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Time at Temperature (m)</td>
<td>19.7</td>
<td>8.0</td>
<td>4.2</td>
<td>12.4</td>
</tr>
<tr>
<td>Flow Rate (LL/Hr)</td>
<td>2.7</td>
<td>1.9</td>
<td>1.2</td>
<td>0.3</td>
</tr>
</tbody>
</table>

**4.4.4 Summary**

The cooling performance of the cold cans was excellent. Using liquid nitrogen, the inner can took 50 minutes to reach a base temperature of 78.4 K, this includes the time to cool down the transfer lines. With liquid helium, the inner can cooled from 94 to 5 K in only 7.5 minutes and responded very rapidly to temperature changes from the cryostat. Temperature stability was observed to be between \(\pm 0.02\) and 0.1 K when using the controller, as was expected to be even better over longer time periods.
Figure 4.4: This plot shows liquid helium cooling data for the cold cans. The initial plateau and peak result from switching the system over from liquid nitrogen. The new base temperature was reached very quickly after switching over to liquid helium. Variations away from base temperature were caused by attempts to determine the minimum cryogen consumption rate and testing the temperature control.
4.5 Recommendations

One area for improvement would be to increase the rate that the outer can changes temperature. The thermal connection between the exhaust shroud of the cryostat and the outer can is already excellent, so there are only two logical parameters that can be adjusted. The first would be to reduce the mass that is being cooled. The front and side panels of the outer can are quite thin, but the solid piece that wraps around the top, back, and bottom could be thinner. For locations with tapped holes, the minimum thickness is defined by how much material is required to hold a tapped 0-80 hole. For everywhere else, the copper can thinned down to the thickness of the panels; that is, $\frac{1}{32}$", or 0.8 mm. With such changes, the mass of the this part could be reduced by 50% or more.

The other change would be to add gold plating to the outside of the inner can and the entire outer can. Maintaining the low emissivity of these surfaces is important for ensuring a low radiative heat load on the inner can.
Chapter 5

The Sample Preparation Stage

In previous chapters, two different solutions for controlling the sample temperature below room temperature were presented. In this chapter, a new apparatus for controlling the sample temperature between room temperature and 1250 °C, the sample preparation stage (SPS), will be described. Unlike previous sample heating stages used by our group, the SPS uses electron bombardment to heat samples. Much of the inspiration for this design came from a 1998 publication by Mendus and Chambers [19]. As with the cold cans, the SPS will be part of the new isothermal STM system currently in development by Drevniok, McLean, and Visser.

The preparation of atomically clean and ordered surfaces is critical to the successful completion of all surface science experiments. To ensure the cleanliness of the sample, microscopy is typically performed at UHV pressures. The UHV pressure range typically includes all pressures below $10^{-9}$ Torr. Any surface under investigation must be cleaned and prepared in UHV to avoid contamination. At UHV pressure, a surface can remain clean for hours or even days. Such conditions allow a prolonged investigation of the prepared surface. In vacuum, sample surfaces can be cleaned
by sputtering and heating. Depending on the composition of the surface, very high temperatures are necessary to produce a clean, well-ordered surface. For example, to clean silicon, temperatures in excess of 1250 °C are required. Accurate temperature control is also necessary for sample preparation. Depending on the sample, long anneal times at a specific temperatures are often required to produce a well-ordered, defect free surface.

5.1 Design Goals and Requirements

The following is a list of the specifications for the SPS:

- All materials must be UHV compatible to temperatures in excess of 1250 °C.
- Sample temperatures of at least 1250 °C must be attainable.
- Temperature control to within ±25 °C must be possible.
- The stage should be designed so that, in the future, tips can be mounted for cleaning.

5.2 Description of the Sample Preparation Stage

5.2.1 Design

Our group’s previous sample heating stages for STM have used resistive heating. This method has worked well because the samples have been almost exclusively silicon. As the new system will also use metal samples, it was decided that electron beam [e-beam] sample heating will be used instead. Electron beam heating works by accelerating
free electrons towards the sample. For the SPS, the electrons were generated by
ermionic emission from a 0.005" diameter tungsten filament. A bias of up to 1000
V accelerated the electrons from the filament to the sample. Upon impingement with
the surface, the kinetic energy of the electrons was converted into heat.

Shown in figure 5.1, the main body of the SPS was machined from stainless steel.
It was designed to mount on the 0.5" diameter shaft from the end of a UHV linear
transfer arm. The entire SPS assembly was designed to fit through the inside of a
2-3/4" CF flange port, which has an inner diameter of 1.5". The entire SPS assembly
without a sample can be seen in figure 5.2.

The chamber and the transfer arm were electrically grounded. In order to bias
the sample, it must be electrically isolated from the rest of the SPS. This was ac-
complished by using alumina spacers and tubes to separate the mounting plate from
the main body of the SPS. Electrical wires for the filament current and sample bias voltage were fixed within alumina tubes at the base of the SPS before making their connections. These wires were bundled together in a fibreglass sheath. At end of the shaft, by the SPS, this bundle was held by a teflon clamp. To allow the transfer arm to extend and retract, the wire bundle was loosely wrapped down the length of the transfer arm. The teflon clamp and a portion of the wrapped wire bundle are shown in figure 5.3.

The sample holders for this system were small plates made of molybdenum. Depending on the shape and type of the sample, different versions of the sample holder could be machined. A small tab on one side of the sample holder interfaced with a tool designed by Will Paul. This tool was used to pick up the sample holder and
transferred it between the SPS and the microscope head. To prevent the sample holder and the mounting plate from welding at high temperatures, the mounting plate was machined from tantalum. Bevels on the front face of the mounting plate guide the sample holder during transfers onto the SPS. Thin tantalum clips were spot welded on the top of the mounting plate. These clips press down on the sample holder to ensure a strong thermal and electrical connection between the sample holder and mounting plate. Figure 5.4 is a photo of the SPS with a sample holder.

To make the filament easier to replace and modify, it was mounted on a removable plate on the bottom of the SPS. This plate and the filament are shown in figure 5.5. One of the filament posts was electrically insulated from the plate by alumina spacers. In this way, a current was passed through the filament by applying a voltage across the two posts. With a sufficient current, Joule heating raised the temperature of
Figure 5.4: The sample preparation stage assembled with a sample and mounted on the transfer arm. A number of important parts have been labelled with the letters A-G: (A) the molybdenum sample plate, (B) the wires for the filament current and sample bias, (C) a tantalum clip to hold the sample plate in place, (D) the tantalum mounting plate, (E) the end of the transfer arm, (F) alumina tubes to anchor and insulate electrical wires, and (G) temporary wires for the k-type thermocouple welded to the sample.
the filament high enough to cause thermionic emission of electrons. Once thermionic emission begins, small changes in current resulted in large changes in the flux of emitted electrons. This effect is mathematically described by the Richardson-Dushman equation, where the current density of the emitted electrons is,

\[ J = AT^2 \exp(-W/kT), \]  

(5.1)

where: \( T \) is the temperature of the metal in K, \( W \) is the work function of the metal in eV, and \( k \) is the Boltzmann constant. Richardson's constant, the last term in equation 5.1, is given by

\[ A = \frac{4\pi mk^2e}{h^3} = 1.20173 \times 10^6 \text{Am}^{-2}\text{K}^{-2}, \]  

(5.2)

where: \( m \) is the mass of an electron, \( e \) is the electronic charge, and \( h \) is Planck's constant.

### 5.3 Testing

The SPS was tested to ensure that sample temperatures of at least 1250 °C could be reached. The sample temperature, the current flowing through the sample, and the sample voltage were all simultaneously collected over the temperature range accessible with the SPS. This data was expected to show a definite, repeatable relationship between the sample temperature and the power applied to the sample. Using this data, we plan to determine the sample temperature without any direct temperature measurements. This would be particularly useful below 450 °C, where pyrometry has proven to be unreliable.

The wire bundle was also stress tested by rotating and translating the magnetic transfer arm to its limits repeatedly. The primary concern was that the wire bundle
Figure 5.5: This is an image of the modular electron-beam heating plate. A 0.005” diameter tungsten filament (A) ran between the two posts on the plate in an ‘S’ shape to provide even heating to the sample. Beneath the filament was a stainless steel plate (B) that mounted to the bottom of the sample preparation stage. This modular design allowed easy access to the filament for repairs and changes.
would either tangle up in itself or bind against the transfer arm. After approximately
50 cycles, including attempts to purposely tangle up the wires, the wire bundle and
teflon clamp were found to remain loose and free.

5.3.1 Experimental Setup

Testing of the SPS was carried out in the preparation chamber for the new STM
system. This was the chamber that it will be installed into and used in when the new
system is fully operational. The preparation chamber was an ideal test location as it
provided the necessary ports for thermometry and power feedthroughs. Additionally,
by testing the SPS in the preparation chamber, very little work would need to be
done to finalise the installation of the SPS after testing was complete.

The bias to the sample was provided by a Glassman\(^1\) MR1P300L power supply.
It was capable of supplying 0.3 A at 1000 V. This power supply could be remotely
controlled by computer, which will be very useful for sample flashes and anneal cy-
cles. For testing, manual voltage control was used. The current for the filament was
supplied by a Kepco\(^2\) CK8-5 regulated DC power supply. The sample current, the
sample voltage, and the filament current were measured using multimeters.

The sample temperature was measured with a k-type thermocouple. The thermo-
couple was directly spot welded to the sample at its junction, as shown in figure 5.4.
Thermocouple temperature was measured by a Fluke\(^3\) 51 K/J Thermometer. K-type
thermocouples are typically only rated up to 1200 °C, as the melting point of Alumel
is 1260 °C\(^4\).

\(^1\)http://www.glassmanhv.com/index.shtml
\(^2\)http://www.kepcopower.com/
\(^3\)http://www.fluke.com/
\(^4\)http://www.matweb.com/search/datasheet.aspx?matid=17350&ckck=1
There were two possible ways to control the power heating the sample. First, the sample bias was held constant while the filament current was varied. Alternatively, the sample bias was varied while the filament current was held constant. Both methods were tested, though the second method was preferred. Fixing the filament current was expected to be less taxing on the filament as its temperature would cycle less often. The filament was the most delicate part and the most likely to fail. Additionally, varying the bias voltage was found to provide finer control over the power applied to the sample.
5.3.2 Results

The SPS with a glowing hot molybdenum sample is shown in figure 5.6. The hottest recorded sample temperature was 1245 °C. When more power was applied to the sample, the thermocouple melted and lost contact with the sample plate. Upon increasing the power, the sample became visibly whiter and brighter. A plot of the sample temperature at a range of applied powers is shown in figure 5.7. The overlap in the data points demonstrated that sample temperature can be reliably extracted from the input power. In figure 5.7, light-grey triangles show the results from varying the sample bias. The black squares and dark-gray circles were the results from varying the filament current.

The data from varying the sample bias was fit by a formula of the form,

\[ T = A \times \ln(P + B) + D \times P + E \times P^2 + F \times P^3 + G \times P^4 + C, \quad (5.3) \]

where T is the sample temperature in Celsius, P is the power in watts, applied to the sample, and A thru G are parameters of the fit. Using Origin’s fitting routines, these parameters were determined and can be seen in table 5.1. The fit, shown in figure 5.8, resulted in \( \chi^2 = 1.14 \).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>310</td>
</tr>
<tr>
<td>B</td>
<td>-3.65</td>
</tr>
<tr>
<td>C</td>
<td>-69.6</td>
</tr>
<tr>
<td>D</td>
<td>-4.18</td>
</tr>
<tr>
<td>E</td>
<td>0.335</td>
</tr>
<tr>
<td>F</td>
<td>-0.00801</td>
</tr>
<tr>
<td>G</td>
<td>((6.418 \pm 0.010) \times 10^{-5})</td>
</tr>
</tbody>
</table>
Figure 5.7: The sample temperature plotted against power applied to the sample, demonstrated the repeatability of the heating and allows sample temperature to be determined from the power.
Figure 5.8: The curve resulting from equation 5.3 with the parameters from 5.1 is shown against the bias varying data. The equation will allow the sample temperature to be calculated based on the power applied to the sample.
Between 20 and 340 °C, the sample temperature was controlled by indirect heating from the filament. In this case, no bias was applied to the sample. The sample temperature is more difficult to control and predict because it takes a very long time for changes in filament temperature to result in a steady state sample temperature.

5.4 Summary

The SPS performed as required and was able to produce sufficient sample temperatures to clean silicon, and hence everything else that will be used in the system. Excellent and repeatable temperature control was demonstrated. With the data collected, the sample temperature can now be predicted and no direct temperature measurement is required. However, if required, a thermocouple could be mounted beneath one of the tantalum clips. Direct optical access to the sample also allows the sample temperature to be determined using optical pyrometry.

5.5 Recommendations

Gathering more data of sample temperature against sample power would be useful. Such data would allow a more accurate prediction of the sample temperature based on the power. When taking these data sets, it is important that the sample temperature is allowed to fully equilibrate before recording it. It is expected that this data will show an even tighter overlap from trial to trial. Along with the thermocouple data, acquiring optical pyrometry data would provide another reference source. Should these data sets correlate well, the pyrometry information would be particularly helpful for temperatures above 1250 °C.
Chapter 6

Conclusion

In this thesis, three different temperature controlling apparatuses for STM were presented and their performance was tested. Each apparatus filled a specific need within our research group. At all stages of a microscopy experiment, controlling, or at least knowing, the temperature of the sample is important. With the cold stage and the cold cans, sample temperature control for microscopy has been extended below room temperature for the first time within our research group. The SPS provides a new level of temperature control and determination for sample heating and preparation.

The cold stage was our group’s first foray into low-temperature STM. It has allowed scanning at temperatures down to 114 K. In addition, the cold stage brought a number of other improvements. Sample transfers are now easier and more robust because of the plunger. The increased thermal contact between the sample puck and cold stage has also resulted in faster cool down times after flashing or annealing the sample.

The cold cans are the cooling solution for the new isothermal STM currently under construction. This new system was designed for low temperature operation from the
The cold cans have been tested with both liquid nitrogen and liquid helium. In both cases excellent temperature control was demonstrated, with a variance of less than 0.1 K away from a set temperature. The inner can of the cold cans has recorded base temperature of 5 K. Temperature control is possible anywhere between 5 K and room temperature.

The final design presented was that of the SPS. The SPS provides the required heating and temperatures to clean and prepare samples of almost any type. Electron beam heating ensures that sample heating is consistent, reproducible, and works with metal samples. Sample temperatures in excess of 1250 °C are possible with the SPS. The temperature can be determined by measuring the power being applied to the sample, which means that neither thermocouples or pyrometry are necessary with the SPS.

It is my hope that each of these innovations will enable exciting, new experiments to be performed by the McLean group here at Queen’s University.
Bibliography


87


Appendix A

Chamber Design for the Isothermal Scanning Tunnelling Microscope

In addition to the work presented in this thesis, two chambers were designed. These chamber are for the isothermal microscopy system being designed and assembled by Drevnoik, McLean, and Visser. These chambers, as well as all of the supporting structures (isolation table, frame, magnetic transfer arms, etc) were modelled using Autodesk Inventor™. This software allowed an accurate representation of the entire system to be created, as shown in figure A.1. The STM chamber and the prep chamber (figure A.1 and A.3), were designed to be as small as possible, while still providing all of the required ports for instrumentation and viewing. Small chambers have the benefit of fast pump-down times and are also easy to handle. Designing such chambers was difficult, requiring a lot of planning, as well as virtual trial-and-error, to find good locations for all of the necessary ports. In both the STM chamber and the prep chamber, the centre point, along the axis defined by the transfer arms, is the focus for a lot of ports. This situation lead to ports being “clustered” together
Figure A.1: This is a rendered image of the isothermal microscopy system. All design work for the isothermal system was performed using Autodesk Inventor™. The cold cans can are located in the STM Chamber [left chamber] and the SPS is located in the preparation chamber [right chamber].

Figure A.2: The SPS can be seen in this image, located within the prep chamber where it will be for sample cleaning and preparation. It is mounted to a magnetic transfer arm that can move it between the STM and prep chambers.
to ensure that they all had line-of-sight to these focal-points. Note, in particular, the cluster of ports at the front of the STM chamber in figure A.3. Many of these ports intersect each other.
Figure A.3: This image shows the isothermal microscopy system. The system is not yet complete. The chamber on the left is the “STM chamber”. The cold cans will be located in this chamber, mounted to a 6” CF flange on the top-most port. The chamber on the right is the “prep chamber”. The SPS is currently installed in the chamber. An image of the SPS in the prep chamber can be see in figure A.2. Sample cleaning and preparation, as well as some tip conditioning, will be carried out in the prep chamber.