μSR STUDIES OF $2H$–$\text{NbSe}_2$

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We accept this thesis as conforming
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Abstract

We report $\mu^+ SR$ measurements in NbSe$_2$ of the penetration depth, $\lambda$, and the core radius, $\rho$, as a function of temperature and magnetic field. We find that $\lambda(H=0T)=1490(11)\text{Å}$ at $T=4.2\text{K}$ and $\rho(H=0T)=248(2)\text{Å}$ at $T=4.2\text{K}$. We observe a field dependence in $1/\lambda^2$ and a strong field dependence in the core radius at fields $H<6\text{kG}$. We also find a temperature dependence in the core radius below $T/T_c=0.3$, where the penetration depth is constant, contrary to Landau-Ginzburg theory. The temperature dependence, however, is much weaker than that predicted by Kramer and Pesch [24], with $\rho(T=0K)=74.6(2)\text{Å}$ in an applied field of $5\text{kG}$. 
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Along with a firm creed in the principles (and benefits) of the scientific method, I believe that science is not a solitary occupation. Instead, it is best and usually only performed by groups of people. Such was the case in the work leading to this Master’s Thesis.

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Chapter 1

Introduction

Although 86 years have passed since Kamerlingh Onnes' discovery of superconductivity, this field continues to yield surprises. The advent of high temperature superconductors in 1986 was one such surprise and sparked an explosion in research in the field. Recently, with the realization that the strong-coupling problem is far from understood, many physicists have returned to take a second look at conventional superconductors, whose properties were eagerly studied until 1986. They have found that conventional superconductors are still not well understood in some areas.

This thesis concerns one of those areas of research, namely the physics of the vortex cores. Vortex cores are tubes of flux that penetrate through type II superconductors. They were predicted by Abrikosov in 1956. Our current understanding of the equilibrium physics of the vortex core is based largely on Ginzburg-Landau theory, complemented by more recent numerical and analytical work using microscopic theories. This thesis presents new data, taken by muon spin rotation, that shows novel behaviour of the fundamental length scales of superconductors, the penetration depth and the coherence length. These measurements indicate that Ginzburg-Landau theory fails to adequately describe the physics of vortex cores below \( T_c/2 \).
2.1 A Superconductor's Properties

The best known and widely quoted property of superconductors is their zero resistivity. Attaching two leads to the surface of superconductor, biasing the contacts, and reading the voltage drop across the leads gives zero resistivity. To show this property, the superconductor must be held at a temperature below a critical value, $T_c$. A more interesting property which distinguishes it from a perfect conductor is a superconductor's ability to completely expel an applied magnetic field, under conditions where the sample is cooled in the presence of the field. This perfect diamagnetism is named the Meissner Effect, in recognition of Meissner who discovered it in 1933.

It is interesting to compare a superconductor with a perfect conductor. A perfect conductor has the same characteristics—low resistance to current flow and magnetic field expulsion. What makes a superconductor different? When an applied magnetic field is increased from zero, to some value $H_c$, a superconductor behaves just like a perfect conductor. According to Maxwell's equations, they both create a magnetisation, $M$, equal and opposite to the applied field, $H$, such that the magnetic field, $B = M + \mu_0 H$, is equal to zero. When the field is reduced from $H_c$, the two materials show different behaviour. The magnetic field will induce current near the surfaces of the superconductor to cancel the magnetic field; the conductor will respond by inducing currents to keep the magnetic field at $H_c$. Furthermore, a conductor may have very low resistance to current
flow, but it will never be zero, as in the case of a superconductor below \( T_c \). In both materials, \( \frac{\partial B}{\partial t} \) is zero, but only in a superconductor is the induced field, \( B \), always zero.

Clearly, given that most conventional superconductors (defined here as having an \( s \)-wave pairing and a phonon mechanism) behave as normal metallic conductors above \( H_c \) and as perfect diamagnets below \( H_c \), there is a phase transition occurring at \( H_c \) and \( T_c \). Furthermore, the transition at \( H_{\text{applied}} = H_c \) is reversible and can be described with the thermodynamic variables magnetic field, pressure, and temperature \( (H, P, T) \).

It is also noticed that when a field is applied, the critical temperature is reduced, and goes to zero when \( H_{\text{applied}} = H_c \). The transition in a finite field is always first order, whereas in zero field it is second order. A simplified version of the phase diagram of a Type I superconductor is presented in Figure 2.1.
2.2 London Model

Under conditions of field cooling, a superconductor expels the applied field. However, the change in induction at the boundary, and the appearance of supercurrents do not occur discontinuously. The induced field, $B$, decays over a length scale $\lambda_L$, introduced in the London model of superconductors in 1935 [2]. They assumed that the current density, $J$, is directly related to the vector potential, $A$. As was shown later using quantum mechanics, this assumption is equivalent to the proposition that the electronic wavefunction in a superconductor is "rigid" or doesn't "bend" in response to the vector potential. In quantum mechanics, the current density in the presence of a vector potential is calculated as

$$\mu_0 \vec{J}(r) = \frac{-ie\hbar}{2m} \cdot (\psi^* \nabla \psi - \text{c.c.}) - \frac{e^2}{mc} \psi^* \psi \vec{A}$$  \hspace{1cm} (2.1)$$

The Londons effectively assumed the first term was identically zero, although their calculations used Maxwell's equations and not the electronic wavefunctions, $\psi$. It should be noted that the relationship between $A$ and $J$ is local; that is the potential at $r$ is determined by the current density at $r$, except for the non-locality introduced by the choice of gauge.

Combining Maxwell's equation

$$\nabla \times \vec{H} = \mu_0 \vec{J}$$  \hspace{1cm} (2.2)$$

with the Londons' assumption yields a simple differential equation for the field and the
current density.

\[
\vec{\nabla}^2 \vec{H} = \frac{e^2 (\psi^* \psi)}{mc} \vec{H}^2 \tag{2.3}
\]

\[
\vec{\nabla}^2 \vec{J} = \frac{e^2 (\psi^* \psi)}{mc} \vec{J}^2 \tag{2.4}
\]

If the magnetic field is assumed to point along the z-axis and the current to flow along the x-axis, then the solution to equation 2.3 is of the form

\[
H(y) = H_0 e^{-y/\lambda_L} \tag{2.5}
\]

where

\[
\frac{1}{\lambda_L^2} = \frac{e^2 (\psi^* \psi)}{mc} \tag{2.6}
\]

and the field is assumed to be \( H_0 \) at the surface \( (y = 0) \) of the superconductor and zero in the bulk \( (y = \infty) \). A similar solution holds for Equation 2.4. The supercurrents and the magnetic field in the superconductor are non-zero only in a thin sheath around the surface of the superconductor. The thickness of the sheath, \( \lambda_L \), is called the London penetration depth. The penetration depth is a property of the material and varies over a wide range in conventional superconductors; for example from 3000Å in Nb alloys to 400Å in Al. It is also a tensor quantity; for example, in the highly anisotropic material NbSe\(_2\), one has \( \lambda_c \ll \lambda \).
A hint that the above scenario of field-induced currents was not quite right came in 1947 in the work of Pippard [3]. He argued that microwave cavity measurements in tin indicated the relation between the induced current and the vector potential was not local. Instead, he proposed a non-local theory of superconductivity, still phenomenological but nevertheless a great advance. He introduced the notion of a Pippard coherence length $\xi_0$, such that the vector potential within a lengthscale $\xi_0$ away from $r$ contributes to the surface supercurrents, in the form of the following equation:

$$ \bar{J}(r) = \int_0^\infty \Gamma(r - r') \bar{A}(r')dr' $$

(2.7)

where

$$ \Gamma(r - r') = e^{-\frac{(r-r')}{\xi_0}} $$

(2.8)

Note that in the London Model

$$ \Gamma(r - r') = \delta(r - r') $$

(2.9)

leading, as noted above, to a local theory.

### 2.3 Ginzburg-Landau Theory

In 1950, Ginzburg and Landau presented their celebrated phenomenological model [4], based on free energies, and the concept of an order parameter, $\phi(r)$. 
They proposed that the transition to a superconducting state at \( T_c \) was characterized by an order parameter. The model was purely phenomenological and only a few parallels can be made with quantities usually present in microscopic theories of condensed matter. The magnitude of the order parameter, \( |\phi(r)| \), can be associated with the density of supercurrent carriers, or electrons, and the square of their wavefunction. The model, however, relies on the assumption that the free energy of the superconductor can be expanded in a series of terms in \( \phi(r) \). The great number of current papers relying on the GL model is homage to the power is of Ginzburg and Landau’s intuition.

The order parameter is complex and decays near the surface of the superconductor or near a vortex core over a length scale \( \xi(T) \), related to Pippard’s coherence length. Inside the bulk of the superconductor, where the superconducting phase dominates, the modulus of \( \phi(r) \) is constant, while its phase can vary with position, indicating the presence of supercurrents. The relation between gradients in phase and current again mirrors the quantum mechanical expression for probability current in terms of the quantum mechanical wavefunction, \( \psi \):

\[
\vec{J} = -i\hbar \cdot (\psi^* \vec{\nabla} \psi - c.c.) \tag{2.10}
\]

As indicated in the discussion on Pippard theory, the order parameter can be twisted or changed in the complex plane over a coherence length. The coherence length is therefore a fundamental quantity of a superconductor, and like the penetration depth, varies greatly in different materials (e.g. \( \xi_0 = 100\text{Å} \) in NbSe\(_2\) and \( 15000\text{Å} \) in Al). An important
point is that the coherence length is often much larger than a typical atomic spacing in most superconducting materials, although it may be on the same order in high temperature superconductors. Discussions of the properties of superconductors on a lengthscale shorter than $\xi$ are not very meaningful in G.L. theory.

2.4 Temperature dependence of $\lambda_{ab}$ and $\xi$

Since $B$ becomes $\mu_0 H_c$ at the critical temperature, $\lambda(T)$ must diverge such that the entire superconductor is penetrated by the magnetic field. G.L. theory yields the critical exponent.

The Ginzburg-Landau theory of superconductivity is based on a free energy written in terms of an order parameter.

$$F_s(T, H) = F_n + \alpha \phi \phi^* + \beta (\phi \phi^*)^2 + \ldots + \frac{1}{2m^*} \hbar^2 \Pi^* \phi^* \Pi \phi + \frac{1}{2\mu_0} \vec{B}^2 - \mu_0 \vec{H} \cdot \vec{M} \quad (2.11)$$

where the set of ellipsis indicate higher orders of the order parameter and $\Pi$ is the canonical momentum in the presence of a vector potential. $\alpha$ and $\beta$ are assumed to have approximate temperature dependences near $T_c$ given by [10]:

$$\alpha(T) = \alpha_0 \left( \frac{T}{T_c} - 1 \right) \quad (2.12)$$

and

$$\beta(T) = \beta_0 \quad (2.13)$$

Looking for the stationary free energy with respect to $\phi$, $\phi^*$, and $A$ leads to:
\[
\frac{1}{2m^*} \hbar^2 \nabla^2 \phi + \alpha \phi + \beta \phi^* \phi = 0 \quad (2.14)
\]

and

\[
\mu_0 J(r) = -\frac{ie\hbar}{2m^*} (\phi^* \nabla \phi - \text{c.c.}) - \frac{e^2}{m^*} \phi^* \phi \mathbf{A} \quad (2.15)
\]

Assuming zero induction and no external currents leads to \(|\phi_0|^2 = -\alpha/\beta\), where \(\phi_0\) is the order parameter deep in the bulk of the superconductor. Equations 2.12 and 2.13 imply:

\[
\frac{1}{\lambda^2} = \phi_0^2 = \frac{\alpha_0}{\beta_0} (1 - \frac{T}{T_c}) \quad (2.16)
\]

near \(T = T_c\) and \(\alpha_0/\beta_0\) is a proportionality constant.

G.L. theory is constructed such that [8, 65] the coherence length has the same temperature dependence as \(\lambda\) near \(T_c\). It can reproduce the familiar decay of the applied field and supercurrent. It is important to note that in G.L. theory, the relationship between the potential and the current is a local one, as in London theory. This may not necessarily hold, as Pippard noted, but the assumption is valid when \(A\) varies slowly on the scale of \(\xi_0\), the Pippard coherence length. A necessary condition is thus:

\[
\lambda(T) = \lambda_L \sqrt{\frac{T}{T_c}} > T - T < \xi_0 \quad (2.17)
\]

Obviously, this occurs when \(\lambda_L \gg \xi_0\) or near \(T \approx T_c\).
It is now clear that much of the physics of a superconductor occurs over the two length scales discussed above: the penetration length and the coherence length. The ratio of the two, defined as \( \kappa = \frac{\lambda}{\xi} \), is another important quantity. It divides all superconductors into two classes, according to:

\[
\kappa > \frac{1}{\sqrt{2}} \quad (2.18)
\]

for type II superconductors and

\[
\kappa < \frac{1}{\sqrt{2}} \quad (2.19)
\]

for type I superconductors.

The division into type I and II superconductors is fundamental, for \( \kappa \) governs how a magnetic field penetrates into a superconductor. \( \kappa \) determines the sign of the energy associated with a wall between a superconducting region and a normal one, as Ginzburg Landau theory shows [10, 8].

We assume that at \( z = 0 \) there is an interface between a superconductor and vacuum and that there is an applied field \( H_{\text{applied}} \). The applied field will decay into the superconductor over a lengthscale \( \lambda \) while the order parameter will increase from zero at the boundary to its bulk value, \( \phi_0 \), over a lengthscale, \( \xi \). The surface energy is given by the difference in the Gibbs free energy of the superconducting state and the normal state, integrated over all \( z \).

\[
\sigma_{ns} = \int_0^\infty dz (G_s - G_n) \quad (2.20)
\]

where the Gibbs free energy is the Legendre transform of the free energy, \( F_s \) of Equation
2.11:

\[ G_s = F_s - \vec{B} \cdot \vec{H} \]  

(2.21)

The boundary conditions are given by:

\[ \phi(z = -\infty) = 0; \quad \phi(z = +\infty) = \phi_0 \]  

(2.22)

and

\[ H(z = -\infty) = H_{\text{applied}}; \quad H(z = +\infty) = 0 \]  

(2.23)

From Equations 2.11, 2.14, 2.15 and the boundary conditions, it can be shown [8, 9] that

\[ \sigma_{ns} = \int_0^\infty dz \left( -\frac{1}{2} \beta \phi^4 + (H - H_c)^2 \right) \]  

(2.24)

Solutions of this integral under the assumption \( \kappa \gg 1 \) give [9]:

\[ \sigma_{ns} \approx -1.104 \lambda H_c^2 < 0 \]  

(2.25)

and that the critical value of \( \kappa \) where the surface energy is zero is given by \( 1/\sqrt{2} \).

2.5 Type II superconductors

From the previous discussion of wall energies, it is clear that in superconductors with \( \kappa > 1/\sqrt{2} \), normal/superconducting interfaces will be favoured and will multiply. It can be shown that in type II field-cooled superconductors, the magnetic field will penetrate into the sample in tubes of flux or vortices, while in a type I superconductor, the magnetic field will only penetrate near the edges of the sample [10]. Cyrot showed that the radius of a vortex was proportional to the coherence length [11].
Since normal/superconducting phase boundaries are energetically favoured, one might ask why vortex cores do not multiply ad infinitum. Two reasons prevent the superconductor from responding in such a way. First, flux is quantized in units of $\phi_0$, because the integral of the phase around a core must be single valued. Furthermore, energy is minimized by having each core carry only one unit of flux. Second, there is a bending energy associated with the curvature of the field as it exits the vortex at the edge of the superconductor. This energy is positive and can be shown to counteract the tendency for vortices to multiply [10].

This picture leads to a modified phase diagram for type II superconductors, shown in
Figure 2.2. The superconductor is in the Meissner state below $H_{cl}$ and at a temperature below $T_c$. In this state all the applied field is expelled except for within a penetration depth of the surface. The bulk of the material is superconducting, characterized by a constant order parameter and phase, in the absence of applied transport currents. As the field is increased to $H_{c1}$, the superconductor experiences a first order phase transition to a state where the applied field penetrates in the form of vortices of flux. As the field is increased further, the number of vortices increases until at $H_{c2}$, they fill the sample and it becomes normal. If the sample is field cooled, the vortices will appear at $T_c(H)$, as the field nucleates into tubes of flux.

2.6 The vortex state

Abrikosov showed in 1956 that the solution to the G.L. equations was a lattice of vortices, with one quantum of flux each. The lattice is depicted in Figure 2.3. He incorrectly calculated that the lattice was square, but later calculations showed that a triangular arrangement of repelling vortices minimizes energy in two dimensions. Vortices in superconductors repel one another because as they approach one another, the magnetic fields in between them add and the intervortex energy of the system increases as the square of the field. Since the force between two vortices is proportional to minus the gradient of the intervortex energy, the vortices repel.

The magnetic field in a triangular lattice is shown in Figure 2.3, part a). The field is maximal at the center of the cores (C), a minimum at the point furthest away from
three cores (A), and reaches a saddle point along the core-core direction (B). The cores are separated by a distance $L$, determined by the average field and the requirement that each cell hold one unit of flux, $\phi_0$. The width of the peak in the distribution shown in Figure 2.4 is proportional to the squared inverse of the penetration depth, $1/\lambda^2$. A large penetration depth therefore yields a narrow field distribution. The tail at higher field in the distribution is due the field in the core of the vortex, and the position of cutoff at high field is governed by the behaviour of the coherence length. A larger coherence length will lead to a cutoff at lower fields, and vice versa.

Abrikosov assumed that the cores were delta functions, with the field rising to infinity inside the cores. Other, more physical, models for the field dependence near the core use Gaussian cutoffs at the center of the core [69, 12] and an analytical solution to the G.L. equations [5].
Figure 2.3: Field distribution in a triangular lattice. Points A, minimum; B, saddle point; C, core.
Figure 2.4: Field distribution with corresponding van Hove singularities labelled as in previous Figure.
Chapter 3

Microscopic Theory and Vortex Core Physics

Ginzburg-Landau (G.L.) and London theory are macroscopic theories, making use of thermodynamic variables and phenomenological parameters. As seen in the case of G.L. theory, the physics they explain are limited in scope by the length scales that enter into the theory.

Yet superconductivity is a microscopic phenomenon where electrons arrange themselves in a particular state to produce very different behaviour in macroscopic observables, such as thermal and electrical conductivity, optical response, etc. New methods such as scanning tunneling microscopy (STM) and $\mu^+SR$ are sensitive to the properties of a superconductor on microscopic length scales and can determine the core radius and the energy levels of bound states. This renewed experimental effort has been matched by a concerted theoretical attack on the problem, based on microscopic theories first elaborated in the sixties.
3.1 BCS Theory

A microscopic explanation of superconductivity was developed in the fifties by Bardeen, Cooper, and Schrieffer, culminating in their celebrated paper, “A Theory of Superconductivity” [32] in 1956.

Earlier measurements of specific heat [80] and ultrasound attenuation [79] indicated that superconductors had an energy gap ($\Delta$) at the Fermi energy. At low temperatures ($kT \ll 2\Delta$), a gap in the density of states at the Fermi energy suppresses thermal excitations. This leads to exponentially activated temperature dependence for many measured properties.

Furthermore, experiments seemed to indicate that electrons were cooperating at a microscopic level to yield the superconducting state. Pippard had guessed at such a mechanism with his theory of non-local current density, but there had been no convincing microscopic explanation of the low temperature properties observed in many superconducting materials.

A breakthrough came when Cooper showed that electrons that are paired in momentum space can form a state that is slightly lower in energy than that of the Fermi gas. For example, in the case of NbSe$_2$ this energy gap is $1.1$meV at $T=0$K[35], compared to a Fermi energy of $35$meV [48].

The final explanation came when Bardeen, Cooper and Schrieffer [14] proposed a groundstate superconducting wavefunction and showed how such a state could have a
lower energy and form a gap in the excitation spectrum at the Fermi surface. They assumed that the attraction between the electrons was mediated by phonons, and that the coupling, which is proportional to the frequency ($\omega$) of the phonon was weak, e.g. ($\omega/\Delta \ll 1$).

The N-particle groundstate wavefunction they proposed was:

$$|\Psi_{BCS} > = \left( \sum_k \frac{v_k}{u_k} c_{k\uparrow}^+ c_{-k\downarrow}^+ \right)^{1/2} |0>$$

(3.26)

where the $c^+$ are electron creation operators and the arrows indicate spin. $u_k$ and $v_k$ are the weighting factors that mix in the vacuum ($u_k$) or hole, and the electron ($v_k$) part of the wavefunction. Superconductivity creates a condensate where all combinations of $u$'s and $v$'s contribute to the wavefunction, each with the same phase. The term quasiparticles, as used in this work, refers to particles that are excited by thermal effects out of the condensate. Their behaviour is similar to nearly-free electrons of normal metals, although in Bogoliubov-de Gennes (BdeG) theory they continue to be described as combinations of states, as in Equation 3.26.

BCS showed that the above wavefunction without spin indices could explain many of the observed phenomena, indicating that the mediating "particle" that pairs the electrons is spinless. This "particle" had long assumed to be the phonon, especially in light of the isotope effect discovered by Frohlich before BCS [17].

It often strikes new students of superconductivity, such as the author, as strange that
two negatively charged particles, namely electrons, could attract one another, even with the help of a phonon. An intuitive and simple explanation is provided by Ziman, which is reproduced here [18]:

"The whole effect springs from a small force of attraction between any two electrons which have nearly the same energy. We usually assume that free electrons repel one another, through their Coulomb interaction.... But in a lattice an electron tends to pull towards itself the positive ions, so that it is surrounded by a region where the lattice is slightly denser than usual. Another electron coming into the vicinity will be drawn towards this region; it will look as if it were attracted towards the first electron."

3.2 Physics of the Cores

In 1989, Hess et al. reported remarkable results of scanning tunneling measurements (STM) of the vortex core in NbSe$_2$[35]. STM measures the tunneling current between the tip of the atomically sharp normal-metal needle, which can be positioned to within an angstrom, and the area directly underneath the tip. Thus only local or bound states, whose wavefunctions are strongly peaked near the tip, are likely to contribute to the tunneling current. Their measurements of the tunneling conductance, which is directly proportional to the local density of states (DOS), showed a non-zero tunneling conductance at the center of the vortex cores.

Their results can be explained in terms of a microscopic picture of the vortex core. Electrons in the vortex state of a type II superconductor can be in two types of states:
scattering (delocalized) and bound (localized) states. Electrons far from the vortex cores tend to be in scattering states and contribute to the diamagnetic currents caused by gradients in the induced magnetic field [48, 44]. Electrons nearer the cores can also be localized or bound within the core. These bound states contribute paramagnetically to the current and are characterized by a quantized energy spectrum. In the isolated core regime, the minigap between bound states is of the order of $\Delta_0^2 / E_F$ and the lowest bound state has an energy $\Delta_0^2 / 2 E_F$, measured from the bottom of the energy gap. This picture emerged from work done by Caroli et al. in 1964 [43, 42].

Kramer and Pesch [23] modified Caroli’s bound state energies in 1974 and found that at low temperatures, the core radius, $\rho_{kp}$, shrinks linearly with decreasing temperature:

$$\rho_{kp} \approx \xi_{kp} = \xi_{BCS} \cdot \frac{T}{T_c} \cdot \left( \frac{3}{2} \right)$$

This is known as the Kramer-Pesch effect.

They used Eilenberger’s semiclassical theory, valid in the region where $T \gg T_c^2 / E_F \approx 130$ mK in NbSe$_2$, and BdeG theory, and showed that both predict the above result. They estimate that the energy gap reaches its BCS value and the current density a maximum at $r = \rho_{kp}$, and that the current density is strongly peaked at $r = \rho_{kp}$, with $J = 5 J_c$. At very low temperatures, the magnetic field forms a cusp at the center of the core and $\Delta_0$ and $J_s$ remain finite. The core remains unchanged below a temperature given roughly by $T = T_c^2 / E_F$. The DOS and the energy of the lowest bound state acquire a logarithmic
correction due to the Kramer-Pesch effect.

\[ N = N_0 2\pi^3 \xi_{BCS}^2 / 3 \ln \left( \frac{\xi_{BCS}}{\xi_{kp}} \right) \]  
\[ \epsilon_{min} \approx \frac{\Delta_{BCS}^2}{2E_F} \ln \left( \frac{\xi_{BCS}}{\xi_{kp}} \right) \]  

For \( T \gg \epsilon_{min} \), the wavefunctions of the quasiparticles vary smoothly on the atomic scale, given by \( k_F^{-1} \approx 15\text{Å} \), while at temperatures comparable to the minimum energy, BdG theory predicts a wavefunction that oscillates rapidly [23, 48] and that \( \Delta_{BCS} \) and \( J_s \) will rise to their maximum over the atomic lengthscale.

Kramer and Pesch calculated the core structure in the presence of very weak core-core interactions, corresponding to very small fields (\( H_{\text{applied}} / H_c \approx 1 \)). However, as has been shown by STM[35] and \( \mu^+ SR \) (see chapter 6 and [70]), core interactions are very important at higher field. Kramer and Pesch suggest that at higher field the core structure changes, but expect the singular behaviour of the field to persist as \( T \) approaches zero [24]. Klein has reported [16] that the effects of a field are to cause the vortex core states to form finite width bands and for the wavefunctions of the quasiparticles to spread out over neighbouring primitive cells. This process is similar to the effect of applying pressure to a non-metallic solid, where under certain circumstances, bound states form bands and the material becomes metallic. Bound states are also predicted above the gap from Bruder's calculations, although the nature of their contribution to the superconductor's
Figure 3.5: Schematic depicting core states. The spacing between the levels is
\[ \Delta \approx \frac{E_{F}^{2}}{E_{F}} \cdot \ln(\xi_{vp}/\xi_{BSC}). \]

electromagnetic response is less clear [41].

After the STM results, Overhauser et.al.[73] offered an explanation for the conductance peak in the center of the cores in terms of electrons hopping from inside the core to outside the core. They assumed semilocalized states in and outside of the core and managed to reproduce the STM results with one parameter. They estimated that the mean hopping rate from inside to outside was given by
\[ t^{2} = \frac{3\pi\Delta}{16N_{0}}, \]
where \( N_{0} \) is the normal density of states at the Fermi surface. It was concluded that the “normal electrons will have self-energy corrections resulting from their coupling (through \( t \)) to the
superconducting excitations outside the core." Their calculations of the self-energy show that the normal electrons with energies $0 < E < \Delta$ have their energies shifted down while those with energies $-\Delta < E < 0$ have their energies shifted up, leading to an increased density of states near $E = 0$ (see Figure 5 of [73]).

Rainer et al.[44], examined the nature of the bound states and reported that the states in the core are predominantly due to Andreev scattering. Andreev scattering involves elastic quasiparticles scattering at a normal/superconducting boundary. The mechanism can be used to explain quasiparticle behaviour at junctions or at core boundaries, where the order parameter rises to its maximum value. In the Andreev scattering process, the quasiparticles scatter off the pair potential ($\Delta_0$), but crucially, reverse charge, group velocity, wave vector, and acquire an effective mass of the opposite sign [41]. In effect, an electron scattering off the order parameter becomes a hole, and vice-versa, with reversed momentum. The coherent superposition of the phases of these scattering events creates a state in the gap[44]. This is in contrast to normal, or specular scattering, where only the momentum component perpendicular to the interface changes sign.

Sipr and Gyorffy [40] point out in their calculations using the BdeG equations that although Andreev scattering contributes most of the states found by STM, normal scattering contributes solely to states of momentum parallel to the core axes. These states have energies above the gap and can only be shown to exist in the context of BdeG theory and not the semiclassical theory.
Earlier work by U. Brandt et.al. [27] confirms the above. They solved the semiclassical equations in high field, investigating the behaviour of the density of states (DOS). Quasiparticles with momentum parallel to the field should contribute to a DOS identical to that of BCS theory with a gap $\Delta_0$, while quasiparticles with momentum in the plane perpendicular to the vortex contribute to a normal metal DOS, i.e. to states within the gap. The effect of a magnetic field is to reduce the gap to zero as $H_{c2}$ is approached.

More recent STM measurements by Hess et.al. resolved more structure in the core [35]. They observe a broadening in the DOS peak in the core as the bias energy is increased. Klein [16] solved the Eilenberger equations with the Green’s function and explained the STM measurements as a shift in the peak that is thermally broadened due to contact between the surface of the sample and a microscope tip that is not at $T=0K$. This was further confirmed by calculations by Machida et.al. [47].

The picture that emerges from BdeG and semiclassical theory and the recent experimental advances is of bound states characterized by a quantum number corresponding to angular momentum. The higher angular momentum states correspond to higher energy states and the wavefunctions of the quasiparticles are concentrated further away from the core (see Fig.4 in [16] or Fig.3 in [49]). This simple physical picture explains the broadened STM peak.

The effects of a magnetic field on this simple picture of core states is unclear at this time. Complications arise due to the non-linearity response of the superconductor near the core [81] and the still unclear contribution to the current density of the bound and
scattering states. However, the behaviour of the bound states should be governed by the reduction in the energy gap caused by a magnetic field.

Finally, we note that all of the above results are calculated in the clean limit. The effects of impurities on the DOS of a superconductor can be dramatic [16]. Impurities fill in the gap in the energy spectrum near $E = 0$ and decrease and broaden the peak in the DOS near $E = 0$. It should be noted, though, that NbSe$_2$ is a very clean superconductor, with $l = 1200\text{Å} \gg \xi_{BCS}$, where $l$ is the mean free path [75, 16].
Chapter 4

Transverse Field $\mu^+ SR$

4.1 Beam properties and muon production

The best muon beams for $\mu SR$ are generated using high intensity proton beams produced at facilities such as TRIUMF. TRIUMF’s 500 MeV cyclotron produces a 140 $\mu$A beam of unpolarized protons by accelerating $H^-$ ions and stripping their electrons with a thin carbon foil. The proton beam is then directed at a production target which feeds secondary muon beamlines.

Both secondary beamlines (M20 and M15) used in this experiment targeted the proton beam at beryllium or carbon targets. The protons interact with the target nucleus, creating pions ($\tau_\pi = 26$ ns) via:

\[ p + p \rightarrow \pi^+ + p + n \]  \hspace{1cm} (4.30)

\[ p + n \rightarrow \pi^+ + n + n \]  \hspace{1cm} (4.31)

which then decay into a positively-charged muon ($\mu^+$) and a neutrino ($\nu$) via:

\[ \pi^+ \rightarrow \mu^+ + \nu_\mu \]  \hspace{1cm} (4.32)

The muon is a charged lepton, with a half life of 2.19709(5) $\mu$s [68]. In the pion rest
frame, the muon has a kinetic energy of 4.12 MeV, which corresponds to a momentum of 29.79 MeV/c.

As a consequence of parity violation [19], neutrinos always have left-handed helicity (spin aligned opposite to momentum), and so the muon must also be left-handed to conserve angular momentum. Nature thus provides us with a ready-made source of nearly 100% spin polarized muons. Such muons coming from pions decaying at rest near the surface of the production target are called “surface” muons.

4.2 Muon interactions in matter

“Surface” muons have a stopping range of about 0.5mm in bulk matter. The fate of stopped negative muons is very different in matter than that of positive muons and we will only be concerned with positive muons ($\mu^+$). The $\mu^+$ decelerates very rapidly and reach thermal energies well within a nanosecond. Because of its low energy and low mass ($m_\mu \approx 200 \times m_e$), the muon creates few Frenkel pairs in the sample compared to heavy ions. Furthermore, its positive charge keeps it a fair distance from atoms and their positive nuclei. It almost always settles at an interstitial lattice position, screened by conduction electrons in metallic samples.

However, the muon does possess a relatively strong magnetic dipole ($\mu_\mu = 1/200\mu_B$) compared to nuclear spins ($\mu_{proton} = 1/658\mu_B$), and thus acts as a very sensitive probe of local magnetism. It will precess in the local magnetic field, $B_{loc}$, at a frequency, $\omega_\mu$, given by:
The gyromagnetic ratio of the muon is [68]:

\[ \gamma_\mu = 135.534(5) \text{MHz/T} \]  

(4.34)

The local field \( B_{\text{loc}} \) will be the sum of contributions from nuclear magnetic dipoles \( (B_n) \), the applied field \( (H_{\text{applied}}) \), and fields generated by supercurrents \( (B_s) \) flowing in the vicinity of the muon site.

\[ B_{\text{loc}} = \mu_0 H_{\text{applied}} + B_s + B_n \]  

(4.35)

We are neglecting other small contributions to the local magnetic field, such as the hyperfine coupling to conduction electrons and the chemical and Pauli paramagnetic shifts.

The muon's energy, mass, charge and dipole moment make it an excellent, relatively inert magnetic probe for use in superconductors [67].

### 4.3 Transverse Field \( \mu^+SR \)

In a transverse field \( \mu^+SR \) experiment one measures the distribution of static local fields in the superconductor. Fluctuations in magnetic field give rise to spin relaxation but the relaxation time in metals, \( T_1 \), is normally much too long compared to the muon lifetime to be observed by \( \mu^+SR \). Typically, the spin polarization of the muons is rotated by 90
deg from both its momentum vector and the applied magnetic field. For a transverse field experiment, the polarization must be rotated using Wien filters for the momentum of the muons to remain parallel to the magnetic field direction. Otherwise the curved muon orbits due to the strong Lorentz force would prevent it from reaching the sample. As discussed above, the muon spin will then precess at a rate proportional to the local field at the muon site. (see Figure 4.6).

The muon spin precession is detected through the parity-violating weak decay of the muon into a positron and two neutrinos (see Figure 4.7).
Figure 4.7: Weak decay of the muon into a positron and two neutrinos. (courtesy of J.H. Brewer)

\[ \mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu \]  

(4.36)

For higher energy positrons, the positron is favoured to decay with its momentum parallel to the spin polarization of the muon at the instant of decay, while for lower energy positrons, the positron is more likely to decay with its momentum antiparallel to the spin polarization of the muon[66] (see Figure 4.8).

However, because the positron energy spectrum is heavily weighted at higher energies, the average asymmetry of the positrons, defined as the number that strike one counter less the number detected in an opposite counter divided by the sum of the detected hits in the two counters, is about one third. Taking into account the fact that the have a finite cross-section and that some of the low energy muons are not detected, the experimental asymmetry is typically about 25%.

To determine the precession rate of the muon, many muon decays need to be detected.
Figure 4.8: Probability of decay positron momentum direction for different positron energies. (courtesy of J.H. Brewer)
Figure 4.9: Counter and sample setup for transverse field $\mu^+SR$. The positron counters are labelled (U)p, (D)own, (L)eft and (R)ight. The outer positron counters are not shown. (courtesy of J.H.Brewer)

Thus, $\mu^+SR$ measures an ensemble of muon decays. The setup of the apparatus is depicted schematically in Figure 4.9.

4.4 Experimental

Measurements between $T = 10K$ and $2K$ on NbSe$_2$ were performed on M20 using the Helios high field $\mu^+SR$ spectrometer with a He gas flow cryostat, and low background insert [20] (see Figure 4.10).

The Helios superconducting magnet can reach fields between 0 and 7$T$. A NMR
probe monitors the field, while a feedback control program controls copper coils around the main magnet which compensate for field drifts. In this manner the field's deviation from the mean is kept below 300mG. A Gaussmeter placed near the NMR probe provided a second reading of the applied field. A gas-flow cryostat bathes the sample in cold He\textsuperscript{4} gas held at a reduced pressure of $\approx 10$ mbar to avoid scattering of muons in the helium gas.

The low background apparatus employs a cup-shaped veto counter that detects muons that miss the sample. It is positioned behind the sample and in the muon beam, and the electronics are wired such that a signal in the veto cup rejects any muon triggering both the muon (TM) and veto (V) counters. The cup shape of the veto scintilator is used to veto decay positrons from muons that miss the sample. This double rejection allows the pileup condition, which rejects events where more than one muon is present in the sample at a time, to be the coincidence of the muon counter (TM) and $\bar{V}$. This greatly increases the rate of "good" muons and reduces what would otherwise be a very large background signal.

Six annular-shaped positron counters positioned in two concentric shells fit into the bore of the solenoid magnet. The inner shell consists of four counters (top left, top right, bottom right, bottom left), while the outer shell consists of two (top and bottom). Requiring a coincidence in the positron signals between both an inner counter and an outer counter defines an event in one of four possible directions (top right, top left, bottom left, and bottom right).
Figure 4.10: Drawing of Helios spectrometer. (courtesy of J.H. Brewer)
Finally, an active collimator can be used in front of the muon counter. The active collimator is a counter with a hole slightly bigger than the beam size (2cm\(^2\) at the muon counter) which vetoes any stray muons or positrons that wander out of the beam.

The electronic logic that defines a start and a stop for the clock is as follows:

\[
\begin{align*}
\text{start} & = TM \cdot \vec{V} \cdot \vec{B} \\
\text{stop} & = E_{\text{top}} \cdot E_{\text{top,left}} \cdot \mu_{\text{gate}} \cdot \vec{V}
\end{align*}
\]

in the case of a positron triggering the top-left counter. \(\mu_{\text{gate}}\) is defined as the coincidence between the muon counter (TM) and a no pileup gate (\(\bar{P}\)). The clock will then increment by one the bin corresponding to the time elapsed between the start and stop in the histogram corresponding to the top-left counter.

In Helios, the sample is mounted on a cryostat insertion rod containing the veto cup. It was attached by a small amount of grease onto aluminized mylar stretched over the veto cup and covered with x-ray mylar, to prevent its loss in case of detachment. The x-ray mylar is thin enough (\(<5\) mg/cm\(^2\)) to avoid any appreciable muon stops. In the present experiment on \(\text{NbSe}_2\), the c-axis of the crystals were aligned parallel to the applied magnetic field.

The average energy of the incoming muons corresponds to a range of 120 mg/cm\(^2\) in carbon. Range straggling of the muon beam is roughly 20-40mg/cm\(^2\), depending on the momentum acceptance of the beamline. The muon counter, cryostat and beamline window, and mylar covering the sample contribute about 53 mg/cm\(^2\) so that the vast
majority of the muons are deeply imbedded in the sample with relatively few stopping upstream of the target. In the case of thin samples, the veto cup rejects those that go through the sample. The temperature was measured with three calibrated diode thermometers, one in the diffuser of the cryostat and the other two near the sample (not shown in Figure 4.10).

The measurements below $T=2K$ were done on M15 in the top-loading Oxford Instruments dilution refrigerator, which has a base temperature of $\approx 10mK$ (see Figure 4.11). The low background requirements made it necessary to modify the way the experiment was done. For example, the typical background signal in a DR measurement is usually about 30% of the total asymmetry, which is unacceptable for this type of lineshape measurement. The higher background in the DR is usually attributed to muons stopping upstream in the sample holder and downstream of the sample in the radiation shields. The problem originates from multiple scattering of the beam in the TM counter, and various windows upstream of the sample. As most of the radiation shielding behind the sample is covered with silver in this area, the signal from the muons will be large, non-relaxing, and at a slightly different field. The field is only homogeneous to 1 part per $10^4$ over a $1cm^3$ volume so a muon 3 cm away from the sample would experience a field 3 Gauss lower than at the sample, in a 1T field. This large background signal would mask the superconducting signal and make detection of effects due to the vortex cores difficult.

This problem was tackled in three ways. Firstly, a veto cup scintillator was installed downstream of the sample and in the muon beam (see Figure 4.11). To do so, windows
Figure 4.11: Drawing of Dilution Refrigerator. Note that the bottom of the veto cup is not shown.
had to be cut into the 50mK, 1K, and 4.2K shields, creating a slightly greater heat load on the DR. The size of these holes could not be too big to keep the heat load through the windows to acceptable levels.

Secondly, as much mass as possible was removed from the beam upstream of the last DR window. This was achieved by removing the back counter and one layer of aluminized mylar foil on the TM counter. In addition, because the stopping density of the sample was relatively small (80mg/cm$^2$ for the thickest piece), two pieces of thin silver foil were wrapped around the sample holder in order to try to range the muons into the NbSe$_2$.

Thirdly, in order to avoid having those muons that went through the sample decay in the silver sample holder, a thin piece of intrinsic GaAs was placed between the sample holder and the NbSe$_2$. Undoped high resistivity GaAs at low temperature does not give a precession signal at the Larmor frequency of the muon as most muons form muonium [78].

The size of the counters was such that they subtended a smaller solid angle from the sample than for the Helios spectrometer. The smaller counters improved the asymmetry but reduced the rate. The vertical design of the DR, with the 1K pot, mixing chamber, and sample transfer lines above the sample, made it impossible to install either an up or down positron telescope. The positron were only detected in two directions, left and right, as opposed to the four directions in the Helios spectrometer. This had the immediate effect of cutting the positron counting rate by roughly half compared to the rate achieved with the Helios spectrometer, as the counters subtended half the cross sectional area from
the sample, and of complicating the fitting procedure for the data, as will be shown in the next chapter.  

Finally, there remained the problem of the heat conductivity through GaAs wafer between the sample holder and the sample. Intrinsic GaAs is an insulator at low temperature, and there was concern that it would prevent cooling of the NbSe$_2$ to 100mK when the only heat conduction is through phonons. The DR’s thermometers were calibrated off-line with a Co$^{60}$ source by nuclear orientation. The tests showed unambiguously that if one waited 90 minutes, the large piece of copper on which the Co$^{60}$ was mounted could be cooled to 15mK from 2K, despite the insulating GaAs that must transfer the heat between the cold finger and the sample (or Co$^{60}$) at low temperatures.

As it turned out, the biggest problem with the DR measurement was the lack of field control. With the Helios spectrometer, the field was controlled to within 300mG with an NMR probe. It was impossible to insert an NMR probe in the bore of the magnet in the DR, and the field could only be monitored with a Gaussmeter, placed 15cm away along a line perpendicular to the solenoid axis. The superconducting Helmholtz magnet on the DR was placed in persistent mode throughout the measurements, but the runs were on average six to seven hours long and flux creep effects changed the field at the center of the magnet by about a Gauss over the course of a run, as measured by $\mu^+SR$. 

4.5 The properties of NbSe₂

The samples of NbSe₂ were made by J.W. Brill at the University of Kentucky. A single crystal of NbSe₂ was used for the experiment on M20, while two different crystals were used in the experiment on M15. A crystal from the same batch as the one used on M20 had a $T_c = 7.0$, while the batch used on M15 had a $T_c = 6.9$. Both had values of $H_{c2}(T = 4.2\text{K}) = 3.5\text{T}$ and $H_{c2}(T = 2.4\text{K}) = 2.5\text{T}$, as measured from magnetisation curves taken with a SQUID.

NbSe₂ is a highly anisotropic layered material. It has a hexagonal layer structure in which each niobium atom has six nearest neighbour selenium atoms. Each selenium atom bonds to three niobium atoms [58]. The Nb-Nb bonds are of a weak van der Waals type while the Nb-Se bond is covalent [57]. The unit cell consists of two layers of Nb-Se. Its c-axis is 12.5Å, while its a-axis is 3.6Å, according to data compiled by Mattheiss [74]. The material is metallic above 7.0K, goes through a charge density wave (CDW) transition near $T=32\text{K}$, and becomes a type II superconductor at $T=7.0\text{K}$.

The charge density wave transition is accompanied by a peak in the magnetic susceptibility, a change in sign of the Hall coefficient, and a change in the splitting of the nuclear quadropole energy levels in NMR measurements. This latter effect indicates that the Nb atoms have moved slightly in response to the CDW, but the movement is too small to see with x-ray diffraction.
de Haas-van Alphen measurements have been made in the normal and superconducting state. They indicate that the Fermi surface of NbSe$_2$ is very small and cylindrical, which allows the CDW to persist in the superconducting state [61]. The niobium 4$d$ bands are responsible for the material’s metallic properties and have an occupied bandwidth of less than 0.7eV. Resistivity measurements indicate that the weakness of the inter-layer bonds lead to a large anisotropy in the electrical properties, e.g. $\frac{\rho_c}{\rho_{ab}} = 31$ at T=300K. This ratio decreases linearly with decreasing temperature [57]. This also leads to a large anisotropy in the penetration depth. In this experiment, we aligned the crystal c-axis with the field, and therefore measured the in-plane penetration depth.

Finally, $\mu^+SR$ measurements by Le et al.[77] have confirmed that NbSe$_2$ is a s-wave superconductor, with no nodes in the gap. Their measurements indicate a 2D-carrier density $n_{2D} \cdot \frac{m^*}{m_e} = 2.9 \times 10^{13} \text{cm}^{-2}$ and a penetration depth of 2500Å. They fitted their $\mu^+SR$ data to a simple Gaussian relaxation function.
Chapter 5

Analysis

The data analysis on superconductors is considerably more complex than in most $\mu^+ SR$ experiments. User (fairly) friendly analysis routines at TRIUMF simplify the task greatly, but extracting physical parameters, as opposed to phenomenological fit parameters, from the data on a superconducting signal is much more difficult.

5.1 The Task

The task entails extracting a superconductor's two length scales, $\xi$ and $\lambda_{ab}$, from the precession signal in two or four detectors. For each counter one records muon decay events (positrons) as a function of time, usually in bins of 1.25 or 0.625 ns, yielding an exponential decay with a time constant equal to the muon lifetime. Furthermore, the spin sweeps by the given counter as it precesses in the field, yielding a small modulation in the counter's data. This oscillation contains all the tranverse field information on the superconductor. The form of the histogram in the $i$th counter is:

$$N_i(t) = N_i(0)e^{-t/\tau}[1 + A_i(t)] + b_i \quad (5.39)$$
Chapter 5. Analysis

where i=1,...,4, \( A_i(t) \) is the positron asymmetry, \( b_i \) is the random uncorrelated background, \( \tau_\mu \) is the muon lifetime, \( N_i \) is a normalization factor and \( N_i(t) \) is the number of events in histogram i in bin t.

5.2 RRF Transformation and packing

After subtracting the random backgrounds, one extracts the asymmetry from two opposing counters. For example, the left and right counter yield an asymmetry given by \( (L-R)/(L+R) \). An assumption is that the asymmetry and normalization factor, \( N_i \), are the same for both counters. This subtraction eliminates the exponential decay and leaves the precession signal. For convenience at higher fields, one usually transforms the signal to a reference frame which is rotating at a frequency close to but below the main frequency of the superconducting signal. Transforming to this rotating reference frame (rrf) involves multiplying the time dependent, complex polarization function, \( P_x(t) + iP_y(t) \), by an imaginary function, \( e^{i\omega_{\text{rrf}}t} \), where \( \omega_{\text{rrf}} \) is the rrf frequency. When there are two pairs of opposing counters, as in Helios, this does not pose a problem, as they can be treated as out of phase with one another by \( \pi/2 \), one pair real and the other imaginary.

However, when there are only two counters, the real part will be present but the imaginary part will not. Multiplication by the exponential will mix the real part into the imaginary and create a high frequency spurious signal close to \( \omega + \omega_{\text{rrf}} \). This problem can be avoided, as was pointed out by Tanya Riseman [68], if the rrf is picked to be an inverse of an integer multiple of the time resolution of the original data. The “created”
imaginary part of the spectrum can then be binned over without adding in any beating
effects.

Once the data has been transformed into the rotating reference frame, it is repacked
to the desired number of data points per unit time. This feature allows the user to
select the number of data points in the plot or to look at different time scales. All of
the NbSe$_2$ data was analyzed over 6 to 7 $\mu$s of data and with a packing such that the
frequency in the rotating reference frame is somewhat greater than the damping rate. A
typical asymmetry signal, transformed to the rrf and packed, is shown in Figure 5.12.

5.3 Fitting the Superconductor $\mu^+$SR Signal

Figure 5.12 contains information on the field distribution in the sample. The damping
rate of the signal will be proportional to the width of the field distribution in the sample,
a quantity directly related to $1/\lambda^2$. A beat can be seen in the signal at certain fields
and temperatures, when the mean superconducting frequency is far enough from the
background frequency (bare muon frequency).

Performing a Fourier Transform of the raw data yields Figure 5.13. The mean width
half height of the signal is proportional to the damping of the signal and to $1/\lambda^2$. The
background is shown at the applied field frequency and sits ontop of the superconducting
signal of the same frequency. A further feature can be seen in the field distribution,
namely the high field cutoff. This contribution is from muons precessing in the cores of
the sample, and is relatively small in weight, compared to the rest of the sample. The
Figure 5.12: NbSe$_2$ signal in rrf: $\omega_{rf}=11.5$MHz, $H=1015$G, T=2.4K.
Figure 5.13: Fourier Transform of $\mu^+SR$ signal of NbSe$_2$ at $H=1885$G and $T=2.4$K.
size of the core radius governs the value of the field in the center of the core. The smaller the core radius, the larger the field at the center of the core, as each core must contain roughly one flux quantum at $H_c^2$.

Fourier Transforms are not, however, the best way to fit data. Ringing effects and the ensuing necessary apodization are consequences of the finite time over which data is taken (see Figure 5.15). Furthermore, information in the error bars is obscured. Instead, the Fourier Transform is best used as a visual check of a feature in the spectrum, for the apodization may change the weight of a feature such as the cutoff, but it cannot shift it to a different frequency. The Fourier Transform also gives an indication of the amplitude of the uncorrelated background, or noise, which determines how long much data must be taken to resolve the cutoff out of the noise.

Because of the aforementioned problems with data analysis in frequency space and because $\mu^+SR$ data is taken in time space, it is better to analyze the data in the time domain. In early experiments, superconducting $\mu^+SR$ signals were fitted with a Gaussian [77]. This method yields only one free parameter, the linewidth, from which one can estimate $\lambda_{ab}$. It is often not a very good fit, as our knowledge of the superconductor’s vortex lattice indicates that there is far more structure in the field distribution than what can be accounted for by a Gaussian.

To solve this problem, Jeff Sonier created a program to fit the signal in time space to a realistic model of the vortex cores proposed by Brandt [13]. A lengthy description of the fitting program can be found in [69]. The program has since been modified to
Figure 5.14: Fourier Transforms at T=2.4K a)H=1.8kG ($\rho = 130\text{Å}$), b)H=4kG ($\rho = 98\text{Å}$), c)H=5kG ($\rho = 79\text{Å}$). Notice the movement of the cutoff.
Figure 5.15: Fourier Transform of $\mu^+SR$ signal of NbSe$_2$ at $H=1015$G and $T=2.4$K. Notice the ringing effect due to the small Gaussian apodization.
Figure 5.16: Fourier Transform of $\mu^+ SR$ signal of NbSe$_2$ at $H=1015$ kG and $T=2.4$ K. Notice that the cutoff is lost in the noise.
fit to a different cutoff proposed by Yaouanc, *et al.* [5]. It fits to nine parameters: the superconducting and background asymmetries, a frequency shift, $1/\lambda^2$, a broadening of the background, $\xi$, the frequency of the background, an additional broadening term due to field inhomogeneities, and a phase. All parameters are allowed to vary freely until the program converges to the lowest $\chi^2$ value.

The theoretical field distribution we fit to is given by [5]:

$$B(r) = \frac{\phi_0}{S} (1 - b^4) \sum_G e^{-\imath \xi \cdot r \over \lambda^2_n G^2} K_1(u)$$  \hspace{1cm} (5.40)

where $u^2 = 2\xi^2 G^2 (1 + b^4)(1 - 2b(1 - b)^2)$, $K_1(u)$ is a modified Bessel function and $G$ is a reciprocal lattice vector of the vortex lattice. $b$ is the reduced field: $b = B/B_{c2}$ and $S$ is the area of the unit cell for a triangular lattice. Figures 5.17 and 5.18 show the induced field $B$ as a function of distance from the core, $r$. It should be noted that the field distribution is rounded near $r=0$, and that the difference between the field at the minimum and at the saddle point is small, even at low fields. According to Yaouanc *et al.*, this field distribution is valid down to $H_{c1}$ and for $\kappa \gg 1$ [5].

Although the program fits to a value we have labelled $\xi$, it is preferable to regard it as a second length scale and not necessarily the coherence length that enters into GL theory. We define the core radius as the distance from the core at which the current density reaches a maximum (see Figure 5.19). Using the field distribution (Equation 5.40) and the fitted values of $\lambda$, $\xi$ and the average field allows us to determine the current density
Figure 5.17: Field distribution along the core-minimum direction (squares) and along the core-saddle point direction (circles). \( H = 500 \text{G}, T = 2.4 \text{K} \).
Figure 5.18: Field distribution along core-minimum direction for different applied fields: \( H = 300 \text{G}, 5 \text{kG}, \) and \( 7 \text{kG}. \) \( T = 2.4 \text{K}. \)
Figure 5.19: Current density profile and core radius. We define the core radius as the distance at which the current density reaches a maximum.

profiles from Maxwell's equation and thereby define the core radius.
Chapter 6

Results

6.1 Measurements of $\lambda_{ab}$ versus field

Because of the relation between $\lambda_{ab}$ and $n_s, n_s \propto 1/\lambda^2$, Figure 6.1 has been plotted, indicating the change in the density of superconducting electrons as a function of field. Both sets of data were fit to a quadratic for the purpose of determining a best fit curve, where $1/\lambda^2 (T = 2.4K) = 58.9(8) - 6.2(1)H + 0.29(2)H^2$ and $1/\lambda^2 (T = 4.2K) = 42.9(1) - 3.87(4)H + 0.087(5)H^2$. The data at $T=4.2K$ are better described by a quadratic, but it must be noted that measurements above 8$kG$ at $T=4.2K$ were not made because of the difficulty of resolving the cutoff in the field distribution at higher temperatures.

Figure 6.2 shows $\lambda$ varying as a function of field. Both sets of data are fit to a line. For $T=2.4K$ (circles) $\lambda = 1300(20)\AA + 71(6)\AA/kG$, and for $T=4.2K$ (triangles) $\lambda = 1490(11)\AA + 100(3)\AA/kG$.

The field dependence of the penetration depth, as indicated in the previous chapter, suggests that the superconducting carrier density is changing with field. A magnetic field is pair breaking [49], meaning that superconductivity dampens a magnetic field or vice versa, as is usual for conventional superconductors.
Figure 6.1: Field dependence of $1/\lambda^2$ at $T=2.4\text{K}$ (circles) and at $T=4.2\text{K}$ (triangles). See text for fitting parameters.
Figure 6.2: Field dependence of the penetration depth at T=2.4K (circles) and at T=4.2K (triangles).
The field reduces the energy gap that the superconducting electrons experience as:

\[ \Delta(H) = \Delta_0 - v_s \cdot p_F \]  

(6.1)

where \( v_s \) is the superfluid velocity and \( p_F \) is the Fermi momentum [15]. The gap disappears when the current density generated by field increases the energy of the electrons by an amount equal to \( \Delta_0 \). Therefore, a higher field will facilitate pair-breaking, as the energy required for the two electrons to leave the superconducting groundstate is reduced. This effect will in turn translate into a field dependence in the equilibrium density of superconducting electrons. This reasoning only holds if one may associate \( 1/\lambda^2 \) with \( n_s \) at higher fields, which is not obviously the case.

The different field dependences of \( 1/\lambda^2 \) at \( T=2.4K \) and \( T=4.2K \) are puzzling. The poor fits of the lowest field points in the \( T=2.4K \) data (see Appendix) make it tempting to suggest that \( 1/\lambda^2 \) depends linearly on field at both temperatures, but the remaining low field data at \( T=2.4K \) still suggest non-linearity. The poor fits are likely due to the small weight of the high field area in the field distribution. At low fields the vortices are well separated and although the core radius is large, the number of muons stopping in the cores is greatly reduced.

It should be noted that the relaxation rate in the normal state also exhibits field dependence, as is shown in Figure 6.3. However, the dependence in the normal state is smaller by two orders of magnitude than in the superconducting state, indicating that the field dependences of \( 1/\lambda^2 \) discussed above is a property of the superconducting state,
Figure 6.3: Gaussian relaxation rate in the normal state of NbSe$_2$ at 10K. The $\mu^+SR$ spectra were fit with a single Gaussian relaxation signal. The normal state relaxation is about $0.2\mu s^{-1}$.

and not effects present in the normal state that persist in the superconducting state.

The origin of this normal-state field dependence in the relaxation rate is still unclear. Inhomogeneity in the magnetic field near the sample would lead to field dependence. An increasing field could also affect the quantisation axis of the Nb nuclear spins, when the Zeeman energy of the $^{77}$Nb becomes greater than the energy due to the gradient in electric field at the Nb site. Neither of these effects, however, would give a linear dependence on field. It is conceivable that the additional broadening is electronic in origin. For example, it could be due to a spread in muon Knight shifts.
6.2 Measurements of $\rho$ versus field

A field dependence for the second length scale in the magnetic field distribution is also found. Figure 6.4 shows that the core radius is strongly field dependent at low field. The data is fit to $\rho = \frac{a}{H} + c$, where $a = 133(3)\text{Å}$ and $b = 0.48(1)$ and $c = 30(2)\text{Å}$ at $T=2.4$K and to a third-order polynomial for $T=4.2$K where $\rho = 248(2) - 58(2)H + 8.5(4)H^2 - 0.47(4)H^3$. Again it should be noted that the $T=4.2$K data does not go above 8kG for reasons mentioned above. Furthermore, the fits are valid only in the region over which the data was taken.
Chapter 6. Results

The change in the core radius as a function of field can be interpreted in a number of ways: macroscopically, an increase in field acts very directly like pressure, due to the quantisation of flux. As the field increases, more vortices enter the sample from the sides (if the sample is cooled in zero field), and the vortices are pushed together. The field is forced higher in magnitude to accommodate the increased flux. Two length scales can vary in this process. One, the penetration depth, increases such that the field decays more slowly away from the cores. The core of the vortex shrinks under the repulsive vortex-vortex interaction, which increases as the cores approach one another.

This reasoning, however, is not very satisfactory. Firstly, it involves macroscopic parameters, such as current densities generating repulsive forces between vortices, which are in turn seen as fairly rigid cores. Secondly, it does not tell us what the core states are doing as the field is increased. A discussion in terms of bound states and the influence of the field on their energies must refer to the work of Kramer and Pesch [23, 26] and Caroli et al. [43, 11]. The calculated energy spacings between the states (minigap) and the energy of the lowest energy bound state above the gap is:

\[ \epsilon_{\text{min}} = \mu \frac{\Delta_0^2}{E_F} \]  

(6.2)

where the logarithmic correction is ignored presently, and \( \mu \) is the angular momentum quantum number of the state (\( \mu_{\text{min}} = 1/2 \)). If the energy gap is reduced by the field, the minigap is also reduced, as it varies as the square of the gap, but the number of states in the gap decreases proportionally to the gap. The lowest bound state energy increases
and the minigap to the next bound state decreases as the field increases. Therefore, all electrons in core states gain in energy as the field is increased.

As the field continues to increase, the higher energy states approach the Fermi energy and electrons are forced by the exclusion principle into the delocalized states with energies above the Fermi energy described by Gygi et.al.[48]. These scattering states contribute to a current that circulates in the opposite sense of the current generated by the bound states [48]. Their increased contribution dampens the current density near the core, which is equivalent to saying that the gradient of the induced field near the core is reduced. The electrons that remain in the core’s bound states lie in low angular momentum states, thereby shrinking the core radius. It should be remembered, however, that only Andreev states can carry current, and that the literature has little to say about their behaviour in changing magnetic fields.

Furthermore, work by Klein[16], Gygi et.al.[48], and Machida et.al.[47] suggest that as the vortex cores approach one another under the influence of the field, electronic wavefunctions in different cores begin to overlap and bands begin to form, leading to the broadening of energy levels familiar to us from band theory. As the field further increases, the band broadening may become equal to the minigap, which itself is shrinking, and lead to a type of continuum of states in the cores below the Fermi energy. This effect is likely to be tempered by the anisotropy that the hexagonal vortex lattice causes in the supercurrent. Gygi and Schluter argue that at higher fields, the star-shaped cores seen in STM [35] force the bound states to quantize angular momentum accordingly, leading
to $\mu = 1/2, 7/2,...$. This effect would increase the minigap.

Another effect that would lead to a shrinking of the cores is that the superconductor becomes less type II as the field is increased (although the nominal value of $\kappa$ increases). The characteristic that separates type I from type II superconductors is the sign of the energy associated with the superconducting/normal phase boundary, as discussed in chapter 4. However, that calculation was made in the single vortex regime, where vortex-vortex interactions were not taken into account. Saint-James et al.\cite{9} choose boundary conditions for the order parameter and vector potential that correspond to a single vortex. This leads to a competition between the order parameter and the magnetic field in the calculation of the free energy of the phase boundary:

$$F_{ns} = \int (|\phi|^4 - B^2)$$

If the effect of neighbouring vortices were included, it is likely that in the regime where vortices are close together, the field term would grow and the proliferation of normal/superconducting boundaries would be less favoured. This effect would lead to a shrinking of the core radius. A proper accounting of the energy associated with the boundary requires a vector potential and order parameter that take account of neighbouring vortices.
6.3 Measurements of $\rho$ at low temperature

Figure 6.5 features a graph of the core radius as a function of temperature. As noted in the figure, two of the points were taken on M20 and show similar linear behaviour, but with a different intercept. The core radius varies linearly with a slope of $-10.6(2) \text{Å}/K$ and an intercept of $74.6(2) \text{Å}$ for the data taken on M15 and with a slope of $-10.61(7) \text{Å}/K$ and an intercept of $66.33(3) \text{Å}$ for the data taken on M20. The value of $\rho$ may be affected by different crystals used on M15 and M20 or by the different apparatus.

We note that previous $\mu^+SR$ measurements of the temperature dependence of the core radius at $H=2\text{kG}$ show a similar temperature dependence for temperatures above $T=2.4\text{K}$ [70]. Figure 6.7 shows the variation in the induced field with respect to distance from the core, along the core-minimum direction on the vortex lattice (see Chapter 3).

The fit for the M15 data did not include the point at $T=100\text{mK}$ because Kramer and Pesch suggest that the core radius no longer shrinks below $T\approx 130\text{mK}$. A fit with the point at 100mK gives a slope of $-9.7(1) \text{Å}/K$ and an intercept of $76.2(1) \text{Å}$. As seen in Figure 6.6, at this temperature, the penetration depth is very weakly temperature dependent, while the relaxation range ($\sigma$) is is not. This is due to the temperature dependence of the core radius, which broadens the linewidth and changes the value of $\sigma$.

The temperature dependence of the core at low temperature is in agreement with the theory of Kramer and Pesch, as discussed in chapter 4. However, the estimates of the core size at low temperature and of the slope are not. Extrapolation of our data
Figure 6.5: Temperature dependence of the vortex core radius at low temperatures. M15 data, triangles; M20 data, circles; point at T=100mK, inverted triangle.
Figure 6.6: Temperature dependence of the Gaussian linewidth at low temperatures (top) and of $1/\lambda^2$ (bottom). M20 data, circles; M15 data, triangles. Notice that $\sigma$ still varies with temperature at $T < T_c/2$, while $1/\lambda^2$ does not, as expected from BCS and GL theory. $T_c(H=5kG) \approx 6.9K$. 
Figure 6.7: Induced field as a function of distance from the core for $T=100\text{mK}$ (inverted triangles); $T=300\text{mK}$ (triangles); $T=500\text{mK}$ (circles); $T=1\text{K}$ (diamonds) and $T=2.4\text{K}$ (squares)
to $T=0K$ give $\rho(T=0K)=74.9\text{Å}$, where the 100mK point is not included in the fit. Taking $\rho_{BCS}$ to be $100\text{Å}$ [16] yields $\epsilon_{min}=3.5 \cdot 10^{-3}\text{meV}$ (the field-induced reduction in the gap for $H=5\text{kG}$ is only 4% and is ignored here). The temperature ($T_{kp}$) at which $\rho_{kp}$ should level off to a constant is $T_{kp} \approx 130\text{mK}$, given a Fermi energy of $35.2\text{meV}$[48], which seems reasonable. Our data does not include points below $T=100\text{mK}$ and therefore we can only say that $T_{kp}$ does not occur above $300\text{mK}$. We suspect that the point at 100mK indicates that $\rho_{kp}$ has leveled off, as predicted above and that $\rho_{kp}(T=T_{kp}) \approx 80\text{Å}$.

We note that this would not alter by much the above estimate of the minigap. However, Kramer and Pesch’s estimates of $\rho_{kp}(T=T_{kp})$ do not agree with our results. They estimate $\rho_{kp} = \frac{\Delta^2}{T_{c}E_F} \rho_{BCS} \approx 1.857\text{Å}$ using again $E_F = 35.2\text{meV}$ and $\Delta_0 = 1.1\text{meV}$.

Thus, the values for the slope and low-temperature core size estimated Kramer and Pesch and those measured by $\mu^+SR$ differ greatly. A number of factors could account for this discrepancy. The theoretical field distribution to which we fitted, described in Chapter 5, does not model the cusp in the magnetic field at the centre of the core that Kramer and Pesch predict. Instead our field distribution shows the field at the centre of the core rising sharply for values of $\rho < 50\text{Å}$. If the magnetic field does have a cusp at low temperature, we would expect our fitting program to overestimate the core radius.

Furthermore, Kramer and Pesch used the isolated-core regime in their derivation. In a field $H=5\text{kG}$, substantial departures from the theory can be expected. It must be noted, however, that the core radius decreases with increasing magnetic field, as shown above, and that $\mu^+SR$ measurements at much lower field would disagree even more strongly
with Kramer et al. ($\mu^+SR$ measurements of $\rho$ near $H_{c1}$ cannot be made due to the small weight of the core in the field distribution: see Chapter 5).

Finally, we note that the energy scales of electrostatic forces lead to the conclusion that cores should be charge-neutral compared to the charge density in the surrounding superconducting bulk. The above calculated value of the minigap, $\epsilon_{\text{min}} = 3.5 \cdot 10^{-3}\text{meV}$, at $T=0K$ indicates that there are approximately 160 states in the gap at $T=0K$. This number of filled states in a core of radius $80\text{Å}$ corresponds to a charge-density of $7.8 \times 10^{13}\text{cm}^{-2}$. However, our $\mu^+SR$ measurements give a 2D charge density of approximately $1.7 \times 10^{15}\text{cm}^{-2}$ (see [77]). Thus either our cores are not charge neutral compared to the superconducting bulk, or our measurements underestimate the number states in the core. The latter is more likely true, as Gygi et al. have already shown that Kramer and Pesch’s linear dependence of the energy of a state on its angular momentum quantum number is not correct for higher energies (see Figure 3 in [48]).
Chapter 7

Conclusions

We have shown novel temperature and field dependence in the fundamental length scales of superconductors. Our measurements of the field dependence of $1/\lambda^2$ indicate unusual behaviour of the electron charge density, while our measurements of the core radius show a strong field dependence at low field, where an increase in the field leads to a rapid shrinking of the cores.

Our low temperature measurements of the core radius indicate that Ginzburg-Landau theory is not valid below $T/T_c = 0.3$, where the core radius continues to shrink in agreement with Kramer and Pesch's theory. However, we do not find the rapid core shrinking that they predict. This is likely due to the fact that their calculations were in the isolated-core regime.

These measurements reveal that conventional superconductors continue to hold surprise for physicists. Further research should be directed to finding the core radius below which the cores no longer shrink, as predicted by Kramer-Pesch theory, and to testing whether the temperature dependence of the core radius is weaker at higher fields, as an
analysis of the behaviour in terms of bound states in the core would seem to suggest. Furthermore, it would also be interesting to investigate the magnetic field dependence of the core radius at lower temperature where there is no appreciable temperature dependence in core size.
Bibliography


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Appendix A: Fits to data

A.1 Data taken in the normal state

The data taken in the normal state was fit with a Gaussian relaxation function, \( e^{\frac{1}{2} \sigma^2 t^2} \).

The \( \chi^2 \) of the fits is shown in Fig A.1.

A.2 Data taken in the mixed state

The data taken in the mixed state was fit with the field distribution described in Chapter 6. Results from the fit are graphed in Figure A.2 for data taken at T=2.4K and in Figure A.3 for data taken at H=5kG.

A measure of the systematic error of experiment can be derived by comparing results taken on the M20 beamline to those on the M15 beamline. This is done in Figure A.4, where the core radius at H=5kG is plotted as a function of temperature. It should be noted that the field in the M20 runs was not exactly the same as the M15 runs, but we believe close enough to give a measure of the systematic error.
Figure A.1: Reduced $\chi^2$ for the fits in the normal state
Figure A.2: Reduced $\chi^2$ as a function of field.
Figure A.3: Reduced $\chi^2$ as a function of temperature.
Figure A.4: Core radius as a function of temperature, at H=5kG. Notice that the runs taken on M20 seem to line on a line with the same slope as in M15, but with a different intercept.