MUON SPIN RELAXATION IN SUPERCONDUCTORS

KEVIN JAKSETIČ

Faculty of Mathematics and Physics
University of Ljubljana

Muon spin relaxation (µSR) is an experimental technique that involves implanting spin-polarized muons into a material, as extremely sensitive probes of magnetism, to measure the magnetic field distribution in its interior. This is achieved by measuring the time-dependent asymmetric angular distribution of emitted positrons or electrons that emerge from the weak decay of implanted muons. Transverse-field µSR is particularly convenient for measuring fundamental properties of type II superconductors, such as their penetration depth. The relaxation of spin-polarization being measured is greatly affected by the formed vortex lattice in such superconductors, which generates a magnetic field distribution inside the sample.

MIONSKA SPINSKA RELAKSACIJA V SUPERPREVODNIKIH

Mionska spinska relaksacija (µSR) je eksperimentalna metoda, pri kateri v material implantiramo spinsko polarizirane mione kot izjemno občutljive sonde magnetizma, da pomerimo porazdelitev magnetnega polja v njem. To dosežemo z meritvijo časovno odvisne asimetrične kotne porazdelitve emitiranih pozitronov ali elektronov, ki izhajajo iz razpada mionov preko šibke interakcije. Mionska spinska relaksacija v transverzalnem polju je še posebej uporabna za določitev nekaterih temeljnih lastnosti superprevodnikov tipa II, kot je na primer vdorna globina. Na merjeno relaksacijo spinske polarizacije močno vpliva tvorjena mreža vrtinčnih niti v takšnih superprevodnikih, iz katere izvira porazdelitev magnetnega polja v vzorcu.

1. Introduction

Muon spin relaxation (µSR) is an experimental technique based on the implantation of spin-polarized muons into a material, to obtain information about its magnetism. It utilizes two key properties of muons: their non-zero spin, which causes them to precess around the direction of the magnetic field, and the fact that muons decay via the weak interaction, with the newborn positrons or electrons being emitted preferentially in the direction of the muon's spin. We will take a closer look at the technique's key features in Section 2.

Superconductivity is a macroscopic quantum phenomenon characterized by ideal conductivity and ideal diamagnetism, first discovered by Onnes in 1911. These properties have sparked significant scientific and technological interest in the study of superconducting materials, including the search for a room-temperature superconductor. After a brief overview of superconducting properties in Section 3, we will finally present µSR as an alternative way of determining the superconducting penetration depth, in contrast to the usual magnetization measurements.

2. Muon spin spectroscopy

Muon spin spectroscopy, more commonly known as muon spin rotation, relaxation or resonance (µSR), is an experimental technique frequently employed in condensed matter research, that involves implanting spin-polarized muons into a material to obtain information about its local magnetism [1].

The muon (μ^-) is a fundamental particle similar to the electron, with an electric charge of $-e_0$ and a spin of $1/2$. As with all elementary particles, there exists its corresponding antiparticle of the same mass and spin but of opposite charge, known as the antimuon or the positive muon (μ^+). Both particles have a mass of $105.66\,\mathrm{MeV}$ [2], which means that they are approximately 200 times heavier than an electron and nine times lighter than a proton. For the purposes of µSR, the negative muon behaves as a ”heavy electron” and likewise the positive muon behaves as a ”light proton” [1, 3].

After being implanted into a sample, both positive and negative muons undergo precession around the direction of the static local magnetic field (which is a sum of the magnetic field generated by the sample and any external magnetic field present), due to their spin. Subsequently, they decay into two neutrinos and either a positron or an electron for positive and negative muons, respectively [1]. By measuring the emitted positrons or electrons, information about the magnetism at the muon's stopping site can be acquired. In the field of condensed matter physics mainly the positive muons are used and so they will be the focus of this seminar.

Muon spin resonance, as an experimental technique in condensed matter physics, is in essence fairly similar to the better-known and more commonly used nuclear magnetic resonance (NMR), but it has some unique advantages over it and can offer new, complementary information about the studied material. Both methods use spin-polarized species as local probes, may that be muons or the material's nuclei, but one of the key virtues of the muon technique is high spin-polarization, which is almost impossible to achieve in NMR. Another important feature of muon spectroscopy is that the experiments can be performed in zero magnetic field, whereas an external magnetic field is necessary for performing a NMR experiment. In contrast to the NMR, which can only be used for samples that are composed of nuclei with magnetic dipoles, µSR can be used for obtaining information on any sample. On the flip side, however, µSR results can be harder to interpret as the position of the implanted muon is usually not known, and due to it being a charge defect, the muon itself could affect the sample's behaviour.

2.1 Muon production

To produce muons in a quantity sufficient for experiments, a proton accelerator is required [1]. Protons are collided into a target made of light elements, usually graphite [3]. The interactions between these nuclei and high energy protons (between 500 MeV and 3 GeV at current facilities) result in pions. Pion production can be summarized as follows:

$$
\begin{aligned}
p + p &\to p + n + \pi^+ \ , \\
p + n &\to p + p + \pi^- \ ,
\end{aligned}
$$

where p and n denote protons and neutrons, respectively [1]. The majority of charged pions decay into a muon of the same charge and a corresponding neutrino with a mean lifetime of 26.033 ns [2],

$$
\begin{aligned}
\pi^+ &\to \mu^+ + \nu_\mu \ , \\
\pi^- &\to \mu^- + \overline{\nu}_\mu \ .
\end{aligned}
$$

These are two-body decays, therefore the muons and neutrinos are emitted with equal and opposite momenta in the rest frame of the pion. Pions are particles with zero spin, whereas muons and muon neutrinos have a spin of 1/2. Consequently, the spin vectors of the newborn muons must be anti-parallel to those of their corresponding neutrinos. Besides this, neutrinos have a definite helicity[1] due to the nature of weak interaction [1, 3]. Only left-handed neutrinos and right-handed antineutrinos exist. Hence, the produced muons are spin-polarized to very high extent. This is one of the key features that enable µSR to be an efficient spectroscopic

[1]Helicity is the projection of a particle's spin onto the direction of its momentum. We say that helicity is right-handed if the direction of the spin is the same as the direction of the motion, and left-handed if the direction of the spin is opposite to the direction of motion.

method [3]. In this article, we will be focusing on the **surface muons**, which originate from the weak decay of low-energy positive pions, and are nearly 100% spin-polarized.

2.2 Muon transport and implantation

Muons possess both charge and spin, which allows us to use a variety of magnets (the beamline) to transport muons from the target to the sample, where they subsequently decay and produce a detectable signal [1].

Positive muons thermalize rapidly when striking matter, over a timescale not exceeding a few nanoseconds. This happens mainly through ionization of atoms and scattering of electrons. Magnetic interactions are not involved, therefore the muons maintain their high spin-polarization [3]. Antimuons implant within interstitial sites in the chemical structure, and can interact with magnetic moments of electrons and surrounding nuclei. Negative muons go through similar processes of losing kinetic energy, though they do not implant in the interstitial sites, but rather close to atomic nuclei [1].

2.3 Muon decay

The muon decay yields a positron (or an electron) and two neutrinos (the mean lifetime of a muon at rest[2] is 2.20 μs [2]), as shown in the following diagram:

$$\mu^+ \to e^+ + \overline{\nu}_\mu + \nu_e \ ,$$
$$\mu^- \to e^- + \nu_\mu + \overline{\nu}_e \ .$$

Since these are three-body decays, the emitted positrons or electrons can obtain a range of energies and momenta. Consider the scenario where we are implanting surface muons (μ^+).

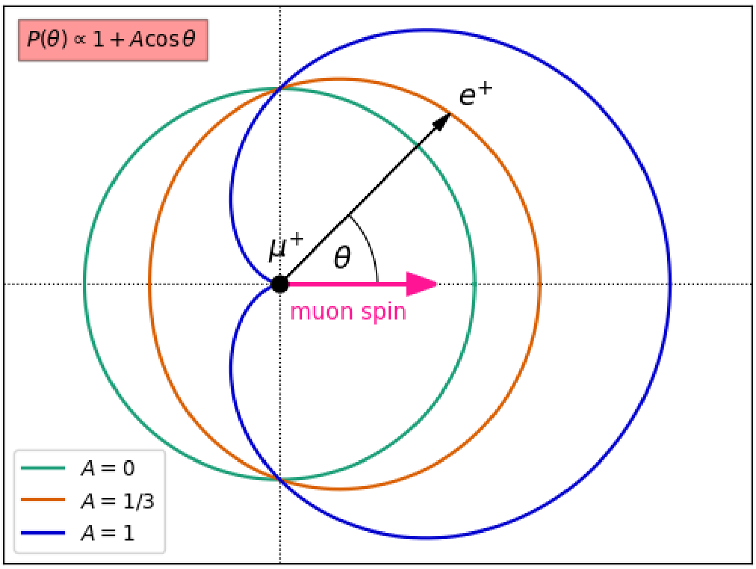

Figure 1. Positron emission angular probability distribution in muon decay. The asymmetry parameter A is equal to 0 for lowest energy positrons and increases monotonically to 1 for highest energy positrons. Its average value is 1/3.

[2] An important advantage of positive muons is that their lifetime is independent of the material in which they are implanted, whereas negative muons captured by nuclei have a material dependent lifetime [3].

They are, in theory, 100% spin-polarized, and the newborn positrons are emitted preferentially in the direction of the muon's spin, with an angular probability distribution

$$P(\theta) \propto 1 + A\cos\theta \ , \tag{1}$$

where θ is the angle between the muon spin and the direction of positron emission, and A is a parameter that describes the asymmetry (that is determined by the nature of the weak interaction) and is dependent on the energy of the emitted positron [4]. Its value increases monotonically from zero to a maximum value of $A = 1$ for the highest energy positrons. If one integrates over all positron energies, the average value turns out to be $A = 1/3$ [3]. Fig. 1 shows how the probability distribution from Eq. (1) changes as A is varied. It is possible to determine the time evolution of the muon polarization by counting the decay positrons (or electrons), and considering that they were emitted preferentially in the direction of the muon's spin [3].

2.4 Experimental setup

The experimental setup is determined by the applied magnetic field, with special interest in zero field, longitudinal field, and transverse field (TF) with respect to the initial muon polarization [1].

A spin vector in a magnetic field precesses around its direction with Larmor precession frequency $\omega_{\mathrm{L}} = \gamma B$, where B is the magnetic flux density and γ denotes the particle's gyromagnetic ratio. For muons it is equal to $\gamma_\mu = 2\pi \cdot 135.5\,\mathrm{MHz/T}$ [1]. This means that we can determine the magnetic field at the muon's stopping site by measuring the precession rate of the spin vector, and considering the angular probability distribution of positron emission from muon decay. The emitted positrons are detected by counters consisting of fast scintillating material and photomultipliers. These counters are placed on opposing sides of the sample to measure the angular asymmetry of the muon decays, utilizing forward-backward, up-down or right-left detectors.

3. Superconductivity

Superconductivity is a macroscopic quantum phenomenon characterized by **ideal conductivity** and **ideal diamagnetism**. As a thermodynamic state, it occurs below a critical temperature T_{c}. Above the critical temperature these properties vanish and the material is in the normal metallic state, in which it has a finite electrical resistivity $\rho > 0$ and it no longer completely expels external magnetic fields from its interior, as shown in Fig. 2.

Superconductors in the superconducting state completely expel an external magnetic field from its interior (providing that it is not too strong). That is why they can be thought of as perfect diamagnets [5]. The magnetic flux density inside a material $\mathbf{B}$ can be written as a sum of the external magnetic field strength $\mathbf{H}$ and the magnetization of the material $\mathbf{M}$ as $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$. In the superconducting state $\mathbf{B} = 0$ holds and consequently $\mathbf{M} = -\mathbf{H}$. Therefore, the susceptibility of a superconducting material in a superconducting state is

$$\chi = \left.\frac{\mathrm{d}M}{\mathrm{d}H}\right|_{H\to 0} = -1 \ .$$

The magnetization is induced by the formation of screening currents on the surface of the superconductor [3, 5, 6]. This expulsion of magnetic flux is called the **Meissner effect**.

It is important to mention that superconductors allow the magnetic field to penetrate their surfaces over a typical length, in the form of exponential decay of the external field,

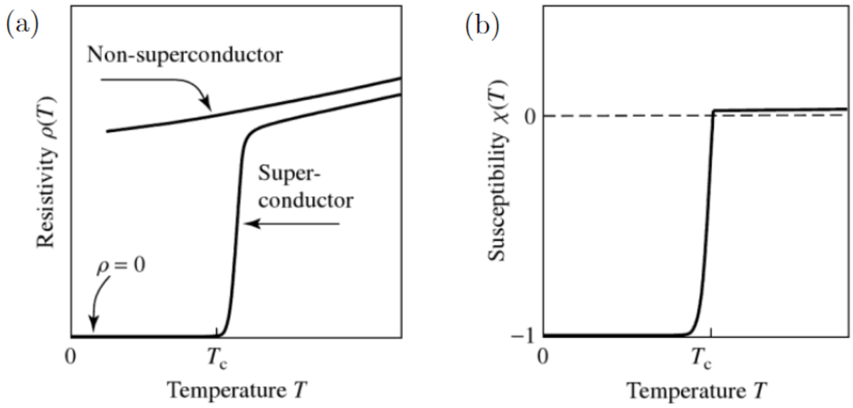

Figure 2. (a) Typical temperature dependence of a superconductor's electrical resistivity, compared to a non-superconducting behavior [6]. A superconductor's resistivity is exactly zero for temperatures below T_c, while a non-superconductor's stays finite. (b) Superconductor's typical susceptibility as a function of temperature [6]. It is exactly equal to -1 below T_c, which corresponds to the magnetic field being completely expelled from the superconductor. Reproduced from [6].

as $B(x) = B_0 e^{-x/\lambda}$. Here, x is the depth inside the superconducting specimen, B_0 is the external magnetic flux density and λ is a parameter known as the **penetration depth**. It can be derived from the phenomenological London theory, which postulates that a fraction of electrons becomes superconducting for temperatures below T_c. We denote the number density of the superconducting electrons by n_s.

There is another superconducting characteristic length ξ, called the **coherence length**. It represents the spatial scale over which significant changes in the density of superconducting electrons can occur, reflecting the fact that it cannot change abruptly [7].

3.1 Type I and type II superconductors

We mentioned that the external magnetic field must not be too strong for the Meissner effect to hold. If the magnetic field strength surpasses the critical value for a given material, the Meissner effect breaks down. We discern between two types of superconductors, based on the way in which penetration of the magnetic field occurs.

For a type I superconductor, there is no penetration of magnetic flux (except on its surface, where it decays over the penetration depth λ) below the critical field $H_c(T)$. This is called the Meissner state. If the applied magnetic field exceeds $H_c(T)$, the entire specimen transitions to the normal state [5]. The typical temperature dependence of the critical field H_c is shown in Fig. 3.

For a type II superconductor, there exist two critical fields: the lower critical field H_{c1} and the upper critical field H_{c2}. Similarly as with type I superconductors, there is no magnetic flux penetration (except on the superconductor's surface) below the lower critical field, as the system is still in the Meissner state. However, when the external magnetic field exceeds H_{c1}, magnetic flux partially penetrates the material in the form of vortices. This state is called the mixed state, or alternatively the vortex state, and it will be discussed more thoroughly in the next subsection. If the magnetic field strength exceeds H_{c2}, the entire sample transitions to the normal state. A typical H-T phase diagram of a type II superconductor is shown in Fig. 3.

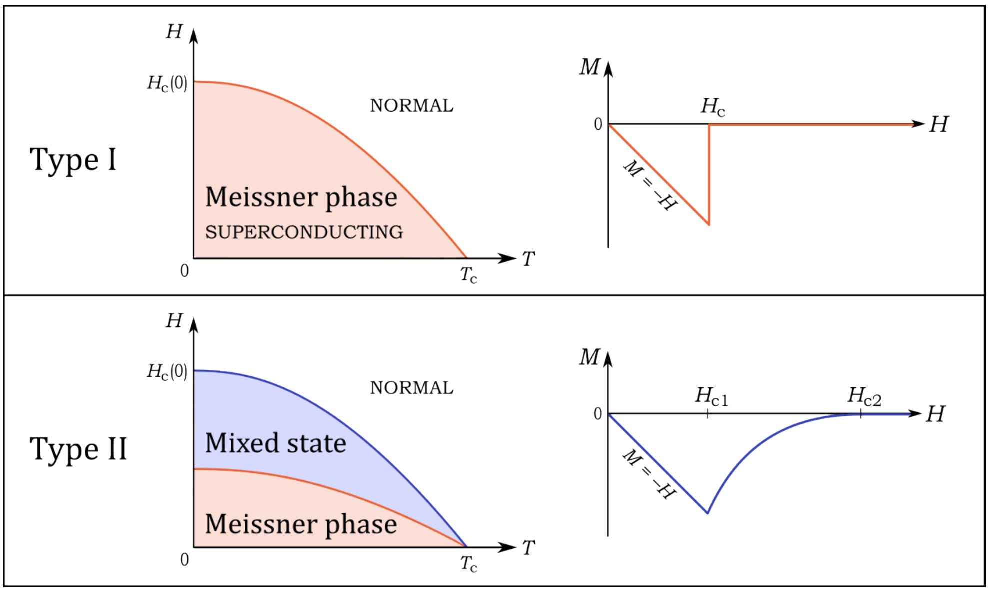

Figure 3. Typical H-T phase diagrams of type I and type II superconductors, with their corresponding magnetizations as functions of the external field H at a constant temperature $T < T_c$. Both Meissner and Mixed state are superconducting, but for a superconductor in the Meissner state, the applied magnetic field only penetrates the superconductor's surface, while it also partially penetrates its interior in the Mixed state. Adapted from [3, 6].

3.2 Properties of the mixed state - the vortex lattice

As mentioned before, type II superconductors exhibit a partial penetration of the magnetic flux in the mixed state. The penetration occurs in the form of flux tubes known as vortices. They are the parts of the material that are in the normal state and usually arrange themselves in a triangular lattice. The spacing between them depends on the strength of the applied magnetic field [7] and is normally in the range of several hundred nanometers, which is orders of magnitude larger than the interatomic spacing [3]. The vortex diameter is proportional to the coherence length and is approximately equal to 2ξ. The magnetic flux flowing through each tube is quantized and equal to the magnetic flux quantum, $\phi_0 = h/2e_0$. Since vortices form additional interfaces between the normal and the superconducting state, superconducting currents arise around the vortices to shield the superconducting part of the material from the magnetic field [3, 7]. Magnetic field density decays over the penetration depth λ, similarly as for the Meissner state [7]. Fig. 4 displays the typical spatial dependence of magnetic flux density, superconducting electron density, and the distribution of supercurrents surrounding the vortices.

In the mixed state, at a fixed external magnetic field, the profile of the magnetic field inside the sample depends on the penetration depth λ and the coherence length ξ, and it has the same translational symmetry as the vortex lattice. In the phenomenological Ginzburg-Landau theory, the so-called **Ginzburg-Landau parameter** $\kappa = \lambda/\xi$ completely determines if a superconductor is a type I or a type II. The theory states that a superconductor is type I if $\kappa < 1/\sqrt{2}$ and type II otherwise, as can be derived using the free energy of the system [3, 7]. When $\kappa < 1/\sqrt{2}$, the formation of interfaces between the normal and superconducting state is not energetically favorable, and the only such interface appears at the material's surface. However, it is energetically favorable for the superconductor to form interfaces for $\kappa > 1/\sqrt{2}$, resulting in the formation of the vortex lattice. A model of the magnetic field distribution of a type II superconductor in the mixed state is shown in Fig. 5.

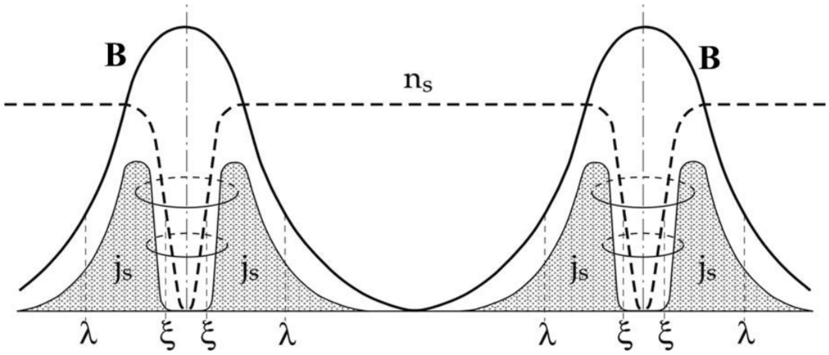

Figure 4. Profiles of magnetic flux density B, the density of superconducting electrons n_s and supercurrents (j_s) around two vortices. Superconducting currents surround vortices in such a way that they screen the external magnetic field, analog to the situation of the Meissner state at the interface between the superconducting state and vacuum. The distance between the vortices is governed by the value of the applied field. Reproduced from [7].

3.3 Measurement of superconductor properties

The penetration depth λ and the coherence length ξ are important properties of a superconductor, therefore we wish to obtain their values for a material of interest. The upper critical field is related to ξ and the lower critical field is related to ξ and λ by the following equations [3]:

$$\mu_0 H_{c2} = \frac{\phi_0}{2\pi\xi^2} \ ,$$
$$\mu_0 H_{c1} = \phi_0 \frac{\ln(\lambda/\xi)}{4\pi\lambda^2} \ .$$

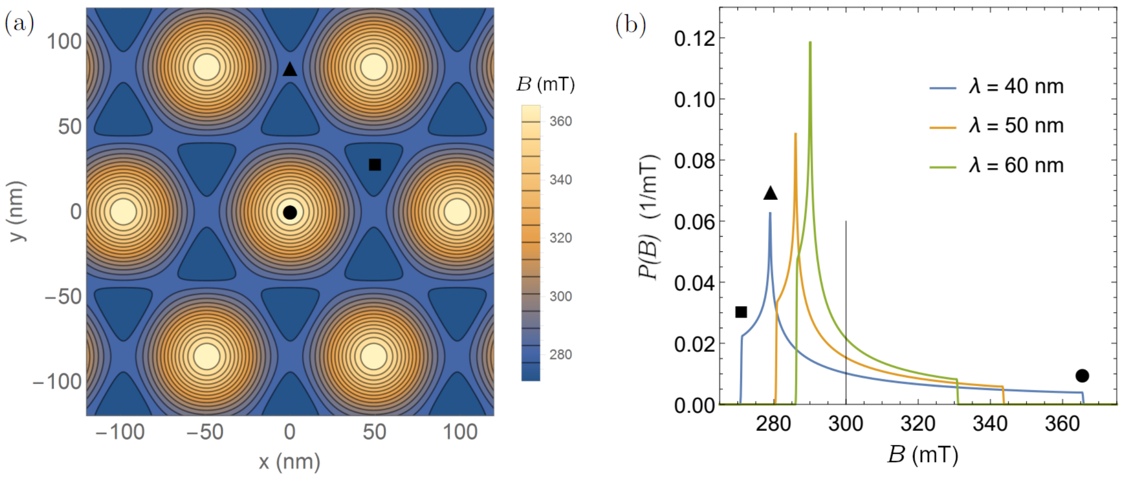

Figure 5. Simulations of the vortex lattice calculated via the London model with a Gaussian cutoff [3]. (a) The magnetic field profile of a sample in the vortex state. (b) Ideal field distribution $P(B)$ in the vortex state for various penetration depths λ. The simulation parameters were $\langle B \rangle = 300\,\mathrm{mT}$ and $\xi = 20\,\mathrm{nm}$ [3]. Adapted from [3].

The upper field H_{c2} can be easily obtained by a magnetization measurement. However, determining the lower field H_{c1} is a much more challenging task, since it is difficult to determine the point at which magnetic flux begins to penetrate the sample. We can use muon spin rotation as a smart alternative that determines the penetration depth λ more directly [3]. The magnetic field distribution can be precisely measured using the µSR technique, if the field variation is sufficiently small on a scale of an interatomic distance, which is indeed the case for fields created by the vortex lattice. The second moment of the magnetic field distribution $\langle \Delta B^2 \rangle = \langle B^2 \rangle - \langle B \rangle^2$ [7] (which represents the standard deviation for a Gaussian distribution of the magnetic field) is related to λ by the equation:

$$\langle \Delta B^2 \rangle = C \frac{\phi_0^2}{\lambda^4} , \tag{2}$$

where C is a parameter that generally depends on the ratio $\langle B \rangle / B_{c2}$ and the Ginzburg-Landau parameter κ [3]. By taking into the account the perfect triangular lattice in the limit $\kappa \gg 1$ (the case of strong type II superconductors), one can find $C = 0.00371$ [7, 3], although the parameter can be calculated for several limits and approximations [7]. We do not know in advance, if the limit $\kappa \gg 1$ holds, so we have to check at the end of the calculation. We will see that it is possible to obtain λ from the µSR technique, and by acquiring the coherence length ξ from a magnetization measurement, we can verify if the limit holds.

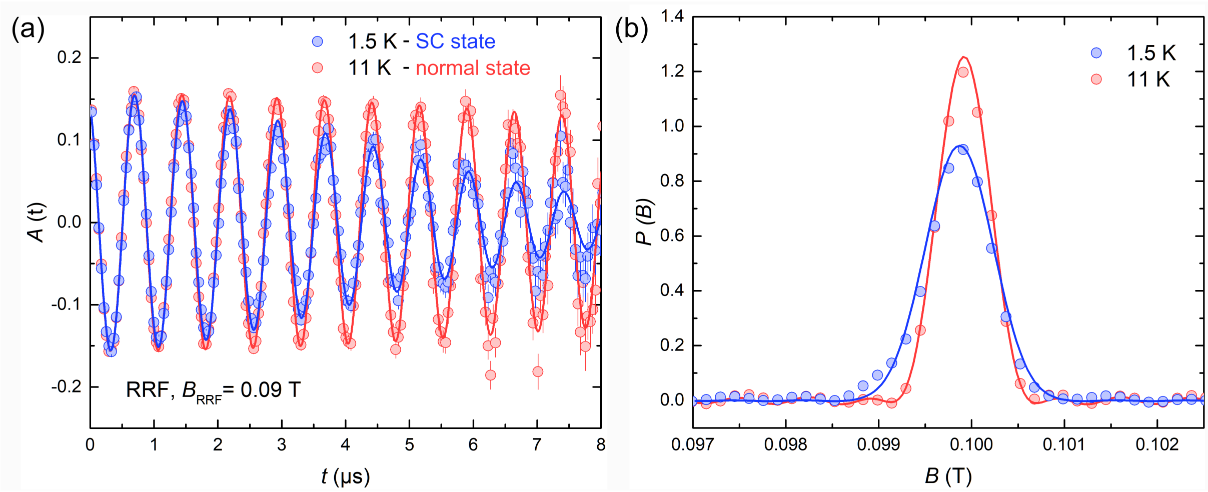

Figure 6. (a) Transverse field measurements of the positron count asymmetry $A(t)$ for a powder sample of $Rb_2Mo_3As_3$, at temperatures $T = 1.5\,K$ (sample is in the Mixed superconducting state) and $T = 11\,K$ (sample is in the normal metallic state), in a field of 0.100 T [8]. Solid red and blue lines are fits to Eq. (3). The results were plotted for a rotating reference frame field $B_{RRF} = 0.09\,T$, which serves us for a better visualization of the data. (b) Fourier transforms of the measured asymmetries $A(t)$ for $T = 1.5\,K$ and $T = 11\,K$. We can see that the normal part of the relaxation is Gaussian. Since the sample was in powder form, the distribution $P(B)$ at $T = 1.5\,K$ is also Gaussian, constructed from a variety of ideal field distributions from Fig. 5. Calculating the standard deviation of the field distribution for the normal state (σ_n) is crucial for obtaining the superconducting part of the relaxation, $\sigma_{sc}(T) = \sqrt{\sigma(T)^2 - \sigma_n^2}$. The sample's penetration depth was calculated to be $\lambda = 669\,nm$ at $T = 1.5\,K$, using Eq. (4). Taking into consideration also the coherence length $\xi = 3.4\,nm$, estimated from H_{c2}, this yields the Ginzburg–Landau parameter $\kappa = \lambda/\xi \approx 200$, which proves that the assumption $\kappa \gg 1$ was valid [8]. Reproduced from [8] or obtained from Quantum Materials Group at the Jožef Stefan Institute.

The magnetic field distribution is obtained via a transverse field µSR experiment, where the asymmetry of positron counts for opposing detectors is being measured (the asymmetry is related to the time-dependent polarization of muons) [8]. In experiments, the field distribution inside a sample is not ideal as shown in Fig. 5b, but looks more like a Gaussian curve as a consequence of various fluctuations and sample defects, especially if one measures a polycrystalline sample [7]. For such samples, the asymmetry can be described using the following formula:

$$A(t) \propto \exp[-(\sigma t)^2/2] \cos(\omega_L t + \varphi) , \tag{3}$$

where σ is the Gaussian relaxation rate, ω_L is the Larmor precession frequency (of the muon) and φ is a phase given by the detector geometries [8]. We can obtain the relaxation rate σ by fitting experimental data to the above equation, Fig. 6. It can be written as $\sigma = \sqrt{\sigma_{sc}^2 + \sigma_n^2}$, where σ_n and σ_{sc} represent the normal and superconducting contributions [8]. The latter is equal to zero above T_c, hence it is simple to determine σ_n (an example is shown in Fig. 6). By assuming a Gaussian field distribution and measuring σ above and below T_c (Fig. 6), one can thus derive σ_{sc}, which directly determines the second moment of the field distribution by the relation $\sigma_{sc}^2 = \gamma_\mu^2 \langle \Delta B^2 \rangle$ [7]. By additionally assuming Eq. (2) with $C = 0.00371$, we find

$$\lambda = 327.5/\sqrt{\sigma_{sc}} \, , \tag{4}$$

where σ_{sc} is in µs^{-1} and λ in nm [7]. Provided that the assumption $\kappa \gg 1$ holds, we have successfully determined λ. The µSR technique is thus an alternative approach for determining the penetration depth of type II superconductors.

4. Conclusion

Muon spin relaxation (µSR) is an experimental technique that implants spin-polarized muons into a material to obtain information about its internal magnetic field, by taking advantage of the known angular asymmetry of the muon decay and measuring the corresponding emitted positrons or electrons. Muons act as extremely sensitive local probes of magnetism and can be exploited to perform measurements of the field distribution inside the samples. This is a vital feature that enables µSR to be a powerful tool for determining important superconductor properties, such as their penetration depth. Transverse field µSR has been widely used to probe the internal magnetic field profiles in the vortex state of type II superconductors [1]. Such samples exhibit an inhomogeneous magnetic field and the depolarization of muon ensembles may be observed. This relaxation is directly connected to the superconductor's penetration depth, from which also the superconductor's lower critical field can be derived when performing an additional magnetization measurement [3]. Thus, muon spectroscopy can be used as a complementary method to NMR and other local techniques. It is not as easily accessible as NMR and the experiments are much more expensive, because of the necessity of having a proton accelerator. Currently there are only a few muon spectroscopy facilities operating around the world, and a couple more are planned, but we can expect that with the ongoing advancements in technology and data analysis techniques, muon spectroscopy will continue to play an important role in the study of superconductors and other complex materials.

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