Muon Study of Novel Quantum States in Thin-Film Superconducting Devices

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Laidlaw Scholars Programme, 2026

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Abstract

This report summarises an internship project investigating novel quantum states in thin-film superconducting devices. The aim of the research is to explore whether a conventional superconductor, niobium (Nb), combined with a material with strong spin-orbit coupling such as platinum (Pt), can generate spin-polarised triplet states and transfer them into a normal metal. Such states could provide a way of carrying magnetic information with much lower energy loss than conventional electronics and are therefore of interest for future superspintronic technologies. A significant part of the project involved developing Python tools for the analysis of SQUID magnetometry and Low-Energy Muon Spin Rotation (LE-μSR) data. The analysis was first implemented for individual muon implantation energies and was then extended to fit several energies simultaneously. This made it possible to obtain information about how the magnetic field changes with depth through the thin film. A more flexible section-based method was also developed to identify possible deviations from conventional superconducting screening without imposing a predefined field profile. The programs were tested using datasets from previous experiments and successfully reproduced measured muon oscillations and produced physically reasonable magnetic field profiles. The next step will be to apply these tools to new measurements from the Paul Scherrer Institut (PSI) to investigate whether spin-polarised quasiparticles can diffuse into an adjacent gold (Au) layer.

1. Scientific Motivation: Towards More Energy-Efficient Computing

Most modern electronic devices, from the phone in your pocket to massive industrial data centres, operate on the principle of moving electrical charge. But this movement is inherently inefficient. Electrons encounter resistance as they travel, meaning they constantly bump into atoms within the material. These collisions cause them to lose kinetic energy that is dissipated as heat, leading to significant energy waste.

As demand for computing continues to grow, managing this heat has become an increasingly important limitation. It limits how small and powerful our devices can be, because maintaining safe operating temperatures becomes increasingly difficult as component density and power increase. The issue extends to environmental concerns, as both computing infrastructure and the cooling systems required to run it consume large and growing amounts of electricity. This has encouraged research into other ways of storing and transferring information.

One promising route is spintronics. While in traditional electronics the basic information carrier is charge, in spintronics information is stored or transported using spin, a different fundamental property. Spin can be loosely visualised by thinking of the electron as a small charge that spins to generate its own magnetic field, so that it resembles a magnetic needle that can point in different directions, like up or down. These different orientations can then be used to represent and transfer information. A second route is superconductivity. Superconductors are materials that, when cooled below a certain temperature, allow electricity to flow with zero resistance and dissipation. This project sits at the intersection of these two approaches, a field known as Superspintronics.

The long-term aim is to understand whether we can use superconductors not only to move charge without resistance, but also to generate, carry and control spin information. The focus of this project is to investigate how we can harness specific quantum interactions in a thin-film superconducting device containing materials like niobium (Nb), a conventional superconductor, and platinum (Pt). Platinum is a normal metal with strong spin-orbit coupling, meaning a strong interaction between the electron's motion and its spin. When a superconductor is layered with a spin-active material, new superconducting states may appear near the interface that allow us to generate long range spin-polarised currents (Flokstra et al., 2023; Lee et al., 2024). We will now look at the quantum mechanics of the interface to understand the basis of this process and the project's methodology.

2. The Quantum Mechanics of the Interface

2.1 Superconductivity and the Meissner Effect

Superconductors not only allow a macroscopic quantum state where electricity flows with zero resistance, they also have unusual magnetic properties. Unlike normal metals, which allow magnetic fields to pass through them, superconductors actively expel internal magnetic fields in a phenomenon known as the Meissner effect. When an external magnetic field is applied, it induces shielding currents on the material's surface that generate an opposing magnetic field. This characteristic of pushing back against an external field is known as diamagnetism. In a superconductor, this diamagnetic screening effectively cancels the external field within the material's interior. This expulsion is not instantaneous at the surface; instead, the magnetic field penetrates a short distance into the material known as the London penetration depth (Blundell et al., 2022). In thin films, fields entering from opposite surfaces can overlap, resulting in a finite internal flux density throughout the sample. In our experiments, we use niobium (Nb), a well characterised conventional superconductor, to establish a baseline state of standard diamagnetic screening. This baseline London penetration depth provides a standard against which to identify the subtle anomalous signals generated by the addition of platinum interfaces.

2.2 Singlets vs. Triplets

In a conventional superconductor like niobium (Nb), electrons do not move through the material as individual particles but form pairs known as Cooper pairs. In these pairs, the electrons exist in a singlet state, meaning their internal magnetic spins are anti-parallel; if one points "up," the other points "down". Because they point in opposite directions, their magnetic orientations cancel each other out, leaving the pair with no net magnetic signal. Since singlet states carry no net spin polarisation, a conventional singlet supercurrent cannot act as a carrier of spin-polarised information.

The devices studied in this project are thin films that consist of layers of material only nanometres thick. This is important because many novel quantum states arise at interfaces, and are not properties of a single bulk material. By layering the superconductor with specific spin-active materials, we can produce triplet states where at least some electron spins are parallel. These triplets are revolutionary because they combine magnetism and superconductivity, exhibiting a unique odd frequency symmetry that allows them to carry a spin-polarised current over long distances without resistance (Eschrig, 2015). This creates a form of novel magnetism that manifests as a partial cancellation of the superconductor's natural diamagnetic screening (Flokstra et al., 2023).

2.3 Spin-Orbit (SO) Interaction

To create these parallel spin-triplets without using traditional permanent magnets, we rely on a phenomenon called spin-orbit (SO) interaction. This is a quantum mechanical effect where an electron’s physical motion through a material is fundamentally coupled to its internal magnetic spin (Eschrig, 2015).

In our research, we use platinum (Pt) because it is a heavy metal with very high SO coupling. When we layer niobium next to platinum, the strong SO interaction at the interface can reorganise the standard singlet pairs from the niobium into the spin-polarised triplet states we need. Furthermore, platinum is a Stoner enhanced paramagnet (Flokstra et al., 2023), a property that helps amplify the magnetic signals to a measurable level.

3. Why Muons?

The main experimental method used in this project is muon spin rotation (μSR). A muon is an unstable subatomic particle that, in our experiments, behaves like an implanted magnetic probe. When a muon lands in a magnetic field, its spin precesses around the direction of the field at a frequency directly proportional to the strength of the local magnetic field at the stopping site, called the Larmor frequency (Blundell et al., 2022).

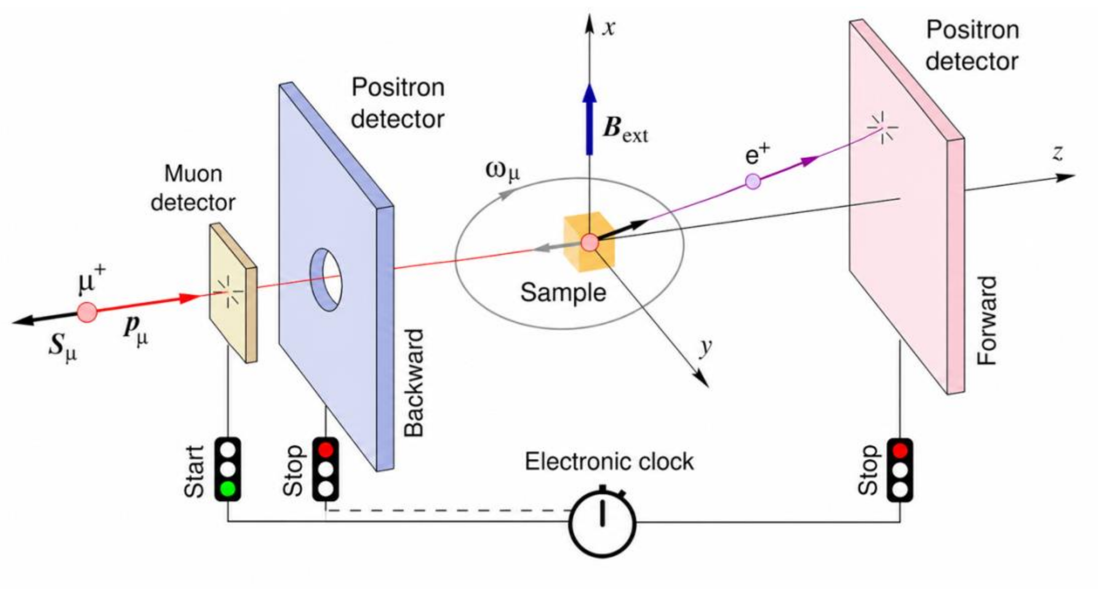

Figure 1. Schematic of μSR experiment. A spin-polarised positive muon is implanted into the sample, where its spin precesses about the local magnetic field. The subsequent positron emission is detected by forward or backward detectors, allowing the time evolution of the muon spin polarisation to be reconstructed. Reproduced from Shang and Shiroka (2021), Fig. 1A, under CC BY.

Muons have a mean lifetime of only 2.2 μs. Positive muons decay into a positron and two neutrinos, with the positron emitted preferentially along the direction of the muon spin at that instant (Blundell et al., 2022; Shang & Shiroka, 2021). Hence, by recording the time and direction of millions of these emitted positrons, we can reconstruct the time evolution of the muon spin polarisation. In a transverse magnetic field, this appears as an oscillating signal; the frequency of the oscillation tells us about the local magnetic field strength, while the damping of the oscillation gives information about the distribution or fluctuations of magnetic fields inside the material.

For this project, we use Low-Energy Muon spin rotation (LE-μSR). Ordinary muons typically implant too deep for our thin-film samples. The Low Energy Muon (LEM) beam at the Paul Scherrer Institut (PSI) can be tuned so that the muons stop at depths of only a few nanometres to a few hundred nanometres, depending on their implantation energy and the material (Morenzoni et al., 2002). The main idea is that changing the muon implantation energy changes the stopping depth; lower energy muons stop close to the surface, while higher energy muons stop deeper inside the sample.

Other than muon techniques, we also use different experimental methods such as SQUID Magnetometry. SQUID stands for Superconducting Quantum Interference Device, and is a tool used to measure the total magnetic moment of a sample. In our research, it provides the necessary preliminary characterisation of the samples and allows us to extract material parameters such as the superconducting transition temperature. However, SQUID magnetometry measures the average response of the entire sample at once, and lacks depth resolution. For our thin-film devices, where the interesting physics occurs at specific interfaces, this bulk averaging is a significant limitation. This is why the depth resolution of LE-μSR is key to this research.

4. Research Context and Objectives

The foundation of this research lies in the team's discovery that placing a conventional superconductor, niobium (Nb), in contact with platinum (Pt) generates an unexpected additional magnetisation near the interface (Lee et al., 2019; Flokstra et al., 2023).

Under normal circumstances, the Meissner effect tells us a superconductor should expel magnetic fields from its interior. However, the experiments revealed a paramagnetic signal (a magnetic response that points in the same direction as the external field) that appeared to partially cancel out the superconductor's natural diamagnetic screening. This phenomenon is attributed to the creation and polarisation of spin-triplet pairs. These triplets are generated through the spin-orbit interactions provided by the platinum (Flokstra et al., 2023). This finding was significant because it showed that we could generate magnetic information within a superconducting state without using traditional ferromagnetic materials, which often cause spin depolarisation and additional energy loss in spintronic devices. In a previous experiment, it was found that the magnetic signal varied with the physical position of the platinum layer (placing it in the middle of the niobium versus an asymmetric position) (Lee et al., 2024). These results supported the idea that we can actively control spin-polarised currents, setting the stage for the project I worked in during my internship.

The current phase of the research looks at moving these spins out of the superconductor and into a normal metal to see if they can be used for information processing. Specifically, during an experimental visit to the Paul Scherrer Institut, we used the LEM beam to investigate whether the spin-polarised quasiparticles generated at the Nb/Pt interface can diffuse into an adjacent layer of gold (Au). Gold was selected as the target metal because of its long electronic mean free path, which allows it to support significant superconducting proximity effects. By proving that spin information can be generated, controlled, and transferred through superconducting states, this research aims to provide the foundation for superspintronic devices that could process vast amounts of data with very low heat dissipation.

5. The Outcomes of the Internship

A major part of my internship was the development of Python-based analytical tools for interpreting magnetic measurements from thin-film superconducting devices. A central challenge of this research is that the experimental data does not directly provide a simple image of the magnetic field inside the sample. My work focused on building programs capable of extracting physical meaning from the data and connecting it to the central question of the project: how does the magnetic field vary inside the sample, and does this variation suggest conventional superconducting screening or additional magnetic effects from spin-triplet states?

The result is a sequence of analysis tools that build on each other. I began with simpler visualisation and single-file fitting programs, and then moved toward more advanced multi-energy fitting programs that reconstruct the magnetic field as a function of depth. I tested these tools with data from previous experiments, and they successfully reproduced the measured muon oscillations and generated physically plausible magnetic field profiles. These initial results demonstrate the potential of the programs for analysing the new experimental data collected during the internship.

5.1 SQUID Data Processing

The first output was a simple program that produces summary plots of SQUID magnetometry data, essentially plotting Magnetisation M against Temperature T or Magnetic Field Strength H. This tool makes the SQUID data easier to inspect and compare, allowing us to move through large sets of measurements.

5.2 Single-Energy Spectrum Fitting

The next stage was to analyse an individual LEM data file. This step allowed me to develop an understanding of what information is contained in a single muon time spectrum before moving to more complex global fits of multiple implantation energies.

In a LEM experiment, spin-polarised muons are implanted into a sample where their spins precess at a Larmor frequency, determined by the local magnetic field. Two positron detectors record the arrival times of decay positrons. As we can see from the expression for the raw counts below, the counts $N(t)$ include several contributions: the intrinsic exponential decay of the muon, the oscillatory signal arising from spin precession, and a time-independent background. The purpose of this single-energy fitting program is to separate these components and isolate the oscillating magnetic signal of interest.

$$N(t) = N_0 e^{-\frac{t}{\tau_\mu}} \left[1 + A e^{-\frac{1}{2}(\sigma\, t)^2} \cos\left(\gamma_\mu B\, t + \varphi\right)\right] + bg$$

In this expression, N_0 represents the initial count normalisation, τ_μ is the average muon lifetime (approximately 2.2μs), and A is the initial asymmetry amplitude. The damping rate σ describes the relaxation of the oscillating signal, which can arise from a distribution of local magnetic fields or from field fluctuations. γ_μ is the muon gyromagnetic ratio ($2\pi \times 135.5$ MHz/T), B is the magnetic field experienced by the muons, φ is the initial phase of the precession, and bg accounts for the time-independent background counts.

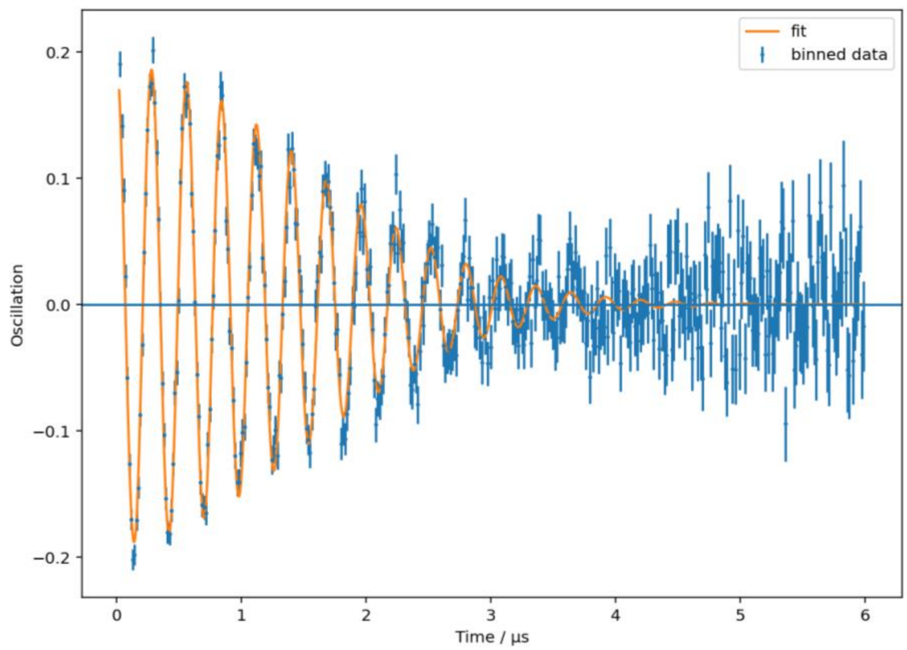

Figure 2. Time-dependent muon spin oscillation signal for a single muon implantation energy. The blue points show the binned experimental data, while the solid orange line represents the best-fit model. The plotted signal isolates the muon spin precession from the exponential muon decay and background contributions, with the oscillation frequency determined by the local magnetic field through $\omega = \gamma_\mu B$

The program assesses the quality of the fit by minimising the chi-squared χ^2 . This statistical concept quantifies the "goodness of fit" by calculating the sum of the squared differences between the experimental data points and the model's predictions. The resulting reduced chi-squared χ^2_{red} , obtained by dividing by the number of degrees of freedom, is then used as an indicator of fit quality. A χ^2_{red} value near 1 indicates that the model accurately describes the data within the limits of statistical noise.

I implemented a two-stage fitting strategy to avoid becoming "stuck" in local minima when changing parameters to minimise the chi-squared χ^2 of the fit. This ensures we find the most reasonable values for the parameters:

1. Global Search: The program first uses SciPy Differential Evolution. This is a stochastic global optimisation algorithm that searches a wide range of parameter values without requiring a manual initial guess.
2. Local Minimisation: Once a starting point is identified through the Global Search, Minuit refines this solution using its MIGRAD variable-metric algorithm and uses the local curvature around the minimum to estimate parameter uncertainties. However, gradient-based optimisers can converge to a local minimum when started from a poor initial guess, which is why the previous step is particularly useful.

This program serves as the foundation for converting LEM measurements from raw detector counts into fitted magnetic information. Its main contribution is providing a tailored μSR fitting tool that doesn't require manual initial guesses, allows users to choose between specific parameters being fitted or fixed, and provides the flexibility to set boundary constraints for each parameter.

5.3 Multi-Energy Field Profile Fitting

After building the single-energy fitting program, the next major step was to find a way to fit multiple implantation energies at once.

We have mentioned that in a conventional superconductor like niobium (Nb), an applied magnetic field is expelled by shielding currents and decays over the London penetration depth. This field decreases approximately exponentially with depth; however, in thin films where the thickness is comparable to the penetration depth, the profile typically follows a cosh-like form as the field penetrates from both surfaces. The introduction of platinum (Pt) interfaces can modify this response; the strong SO interactions in Pt reconfigure standard spin singlet pairs into spin-triplet correlations, manifesting as an additional paramagnetic contribution that opposes standard screening. To determine whether these novel states are present, we use Low Energy Muon Spin Rotation (LE-μSR) as a depth resolved magnetic probe. By mapping the field profile on a nanometre scale, we can determine whether the data is consistent with ordinary superconductivity or if extra features, such as triplet induced magnetism, are needed to describe the system.

A central challenge in LE-μSR is that muons do not stop at a single depth. Instead, each implantation energy has a corresponding stopping distribution; a range of depths over which the muons are likely to stop. These stopping profiles broaden and shift deeper as implantation energy increases, and are simulated using Monte Carlo methods such as TRIM.SP. Consequently, the experiment does not measure the field at one specific point, but a weighted average over a range of depths, effectively hiding the underlying local magnetic field profile B(x) within the raw data.

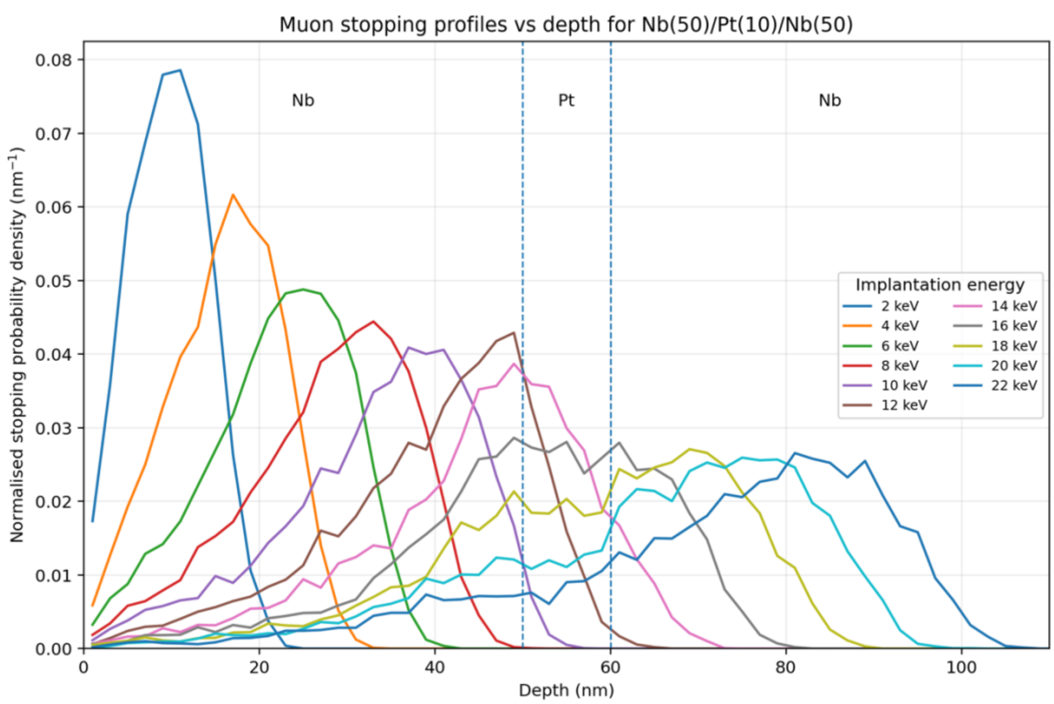

Figure 3. Normalised muon stopping probability density as a function of depth for implantation energies from 2 to 22 keV, obtained via Monte Carlo TRIM.SP simulations for the multilayer Nb(50 nm)/Pt(10 nm)/Nb(50 nm). The profiles show broadening and shift deeper into the sample as implantation energy increases.

Before this project, the different implantation energies were primarily inspected through an iterative manual procedure. I implemented a global fitting framework that fits the full set of implantation energies simultaneously; it assumes one shared depth-dependent field profile B(x) and calculates the expected muon signal for each energy by weighting that profile according to the corresponding stopping distribution. This represents a significant advancement over the single-energy fitting program, which could only provide depth averaged field values and not a continuous spatial variation.

A useful feature of the program is that the magnetic profile is written as a plug-in function, so that the rest of the analysis does not need to be rewritten to test different models. The idea is to start by testing a cosh-like superconducting screening function consistent with ordinary superconductivity and compare the fit to that of more complex physically motivated profiles.

The program produces plots of the best fit magnetic field profile B(x), the stopping profiles used in the fit, and isolated oscillation plots with fitted curves for every detector and energy. This tool represents one of the central outputs of the internship, as it converts a collection of separate LEM files into a unified physical model of the sample's magnetic response.

5.4 Free-Form Field Profile Reconstruction

The most advanced tool I developed during this project moved beyond assuming a fixed magnetic field shape. While model based functions, such as a cosh profile, are very effective in testing particular hypotheses, they also impose a particular physical form on the results. Since the core objective of the project is to search for possible anomalous magnetic behaviour, it is important to have a method that does not bias the data toward a conventional description too early in the analysis.

To address this, I developed a program that reconstructs the magnetic field profile in a model-independent way. Rather than assuming a global mathematical function, the program divides the thickness of the film into discrete depth sections and optimises an independent magnetic field value for each depth section. This produces a step-like profile, where each section is free to take its own value. The program still fits all implantation energies simultaneously, but instead of using a single global formula for the magnetic field, it finds the specific set of section fields that collectively provides the best match for all measured muon oscillations.

This tool allows the data to suggest the shape of the magnetic profile before a specific model is imposed. A conventional cosh profile can be overlaid afterwards as a comparison, rather than being built into the analysis from the beginning.

The reconstruction has finite depth resolution because each implantation energy samples a broad stopping distribution; neighbouring section fields may therefore be correlated; a highly flexible profile can reproduce statistical noise; and the reconstructed profile needs to be evaluated using parameter uncertainties and model comparison before deviations can be interpreted as evidence of anomalous magnetism.

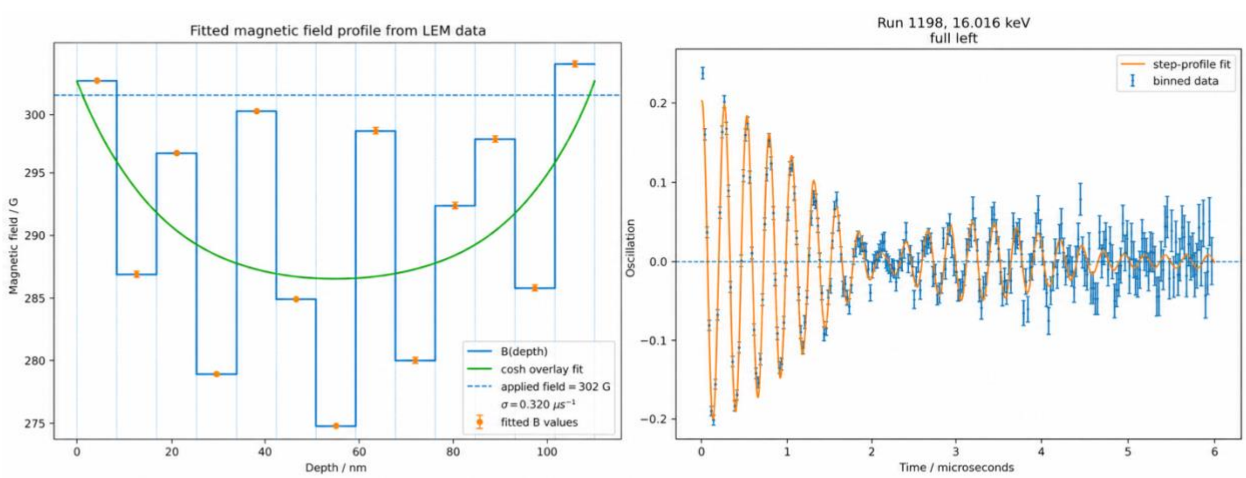

Figure 4(a) and Figure 4(b). Left: reconstruction of the magnetic field profile (B(x)) across the film thickness, showing discrete step-like depth sections. The green curve shows a conventional superconducting screening cosh-like profile for comparison, highlighting deviations from conventional screening behaviour. Right: corresponding time-dependent muon spin oscillation signal at an implantation energy of 16.016 keV. The blue binned data show the experimental oscillatory signal, while the solid orange curve shows the signal predicted from the fitted step-profile in Figure 4(a).

6. Conclusion

This internship project has contributed to the ongoing study of spin-polarised triplet states in superconducting thin-film devices. The primary achievement was the development of Python-based analytical tools to interpret SQUID and LEM magnetic measurements. Prior to the development of these tools, the analysis of the data heavily relied on a manual, iterative fitting process. This approach was useful for testing specific hypotheses, but it has important limitations when used on its own.

The programs developed allow for a more exploratory, data-driven approach; in particular, the Free-Form Field Profile Reconstruction program works as a free section field reconstruction tool that doesn't assume a fixed shape for the magnetic profile. Furthermore, working out how to fit multiple implantation energies simultaneously ensures that a single, unified physical model accounts for the weighted spatial averages measured at every depth, enforcing self-consistency across the data set. Incorporating fitting tools also improves efficiency and flexibility. Manual fitting is time consuming, especially when many implantation energies, detector pairs, and stopping profiles must be considered. Much of this process can now be automated while making it easy to make controlled fitting choices, such as setting boundaries or fixing parameters. The programs were tested using previous experimental datasets, successfully reproducing measured muon oscillations and generating physically plausible depth-dependent magnetic field profiles. They proved useful not only for validating specific theoretical models, but also for pointing out unexpected characteristics that could otherwise go unnoticed.

Moving forward, the immediate next step is to use these tools to analyse the experimental data collected during the recent beamtime at the Paul Scherrer Institut (PSI). This analysis will focus on investigating whether spin-polarised quasiparticles generated at the niobium/platinum (Nb/Pt) interface successfully diffuse into an adjacent gold (Au) layer, which was selected as a target metal due to its long electronic mean free path. Showing that these spin-polarised states can be generated, controlled, and transferred into a normal metal would help provide the scientific foundation for potential future superspintronic devices capable of high-density data processing with minimal energy dissipation.

Acknowledgements

I would like to thank Stephen Lee for his guidance and support throughout this project, and the wider research group for their advice and discussions. I am also grateful to the staff at the Paul Scherrer Institut for their assistance during the LEM beamtime. Finally, I would like to thank Lord Laidlaw and the Laidlaw Foundation for supporting this internship.

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