Hyperfine Magnetic Fields in Fe/Ag Magnetic Multilayers Probed with Low Energy Spin Polarized $^8$Li

by

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This thesis is a presentation of experiments examining the induced hyperfine magnetism in the nonmagnetic layers of two thin film magnetic multilayers, Au(40Å)/Ag(200Å)/Fe(140Å) and Au(40Å)/Ag(800Å)/Fe(20Å) grown on GaAs(001) single crystal substrates. The main technique used in this study was beta-detected nuclear magnetic resonance (βNMR) conducted at TRIUMF laboratories in Vancouver, British Columbia. βNMR makes NMR measurements on radioactive $^8$Li$^+$ ($\tau=1.21$ sec) nuclei that are implanted directly into the sample. Resonant βNMR experiments showed strong induced magnetism in the non-magnetic Ag layer due the magnetic Fe. Comparison of a theoretical model to experimental results on the Au(40Å)/Ag(200Å)/Fe(140Å) sample suggest that the induced hyperfine magnetism decays with distance into the Ag layer from the magnetic/nonmagnetic interface like $x^{-1.84}$ from a maximum of $\sim0.3$ T at the magnetic/nonmagnetic interface.
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Chapter 1

Introduction

We assume the matter our macroscopic world is comprised of will always display the properties we observe in our everyday life. For example, if one were to cut a block of some material in half, we expect each piece to exhibit the same properties that the whole did. If one were to repeat this procedure over and over, however, eventually one would be left with a piece so small that any further division would change the material’s intrinsic properties. This is the boundary between the macroscopic world of our existence and the nanoscopic world, where length is measured in nanometers (billionth of a meter), of atoms and molecules. The approximation that any atom is surrounded by an infinite number of other atoms in all directions is no longer valid because on the scale of nanometers the material will only be made up of a few atoms. The “bulk” properties of the material are reduced, and the “surface” properties become much more important, since on average an atom is more likely to be near the surface as the total number of atoms is reduced. We can engineer structures in a way that exploits this, and use it to study specific effects in ultra small scale systems. By growing “ultra thin films” only several atoms thick, we have effectively reduced the number of dimensions of this system from three to two where nearly every atom is at the surface and surrounded by many atoms on all sides but very few above or below.

The effect of low-dimensionality on magnetic materials is particularly striking. Ultra-thin magnetic films act like giant magnetic molecules with their own unique magnetic properties [1]. In 1986 Grunberg et al. [2] made the unusual discovery that when two ultra thin films of Fe were separated by a thin Cr layer of certain thickness, the magnetization direction of the Fe films align antiferromagnetically in the absence of any external magnetic field. This led to the independent discoveries in 1988 by Fert and Grunberg [3, 4] that the resistivity across these layers would decrease dramatically when the coupling of the Fe layers changed from antiferromagnetic (AF) to ferromagnetic (FM). This effect, which has been attributed to spin dependent
scattering of conduction electrons at the magnetic/nonmagnetic interfaces, is much larger than the conventional magneto-resistance described by the Lorentz force, and was thus given the name giant magneto-resistance (GMR). This phenomenon was quickly adopted for use hard disk read heads, greatly increasing their sensitivity, thus enabling the rapid increase seen in hard disk bit density. In 1990 it was found that interlayer exchange coupling (IEC) of magnetic layers oscillates as a function of spacer thickness for certain magnetic multilayer (MML) structures [5, 6, 7], later this phenomenon was shown to occur in almost any system of magnetic layers separated by a transition metal spacer, and that they all oscillate with approximately the same period, \(\sim 10 \, \text{Å} \) [8]. These magnetic structures hold great promise in the burgeoning spintronics industry which involves engineering electronic devices, such as spin transistors and magnetic random access memory (MRAM), which utilize electron spin as well as charge by specifically tailoring of a material’s magnetic and electric properties.

This oscillation between FM and AF coupling of the magnetization in magnetic thin films seems counterintuitive based on our everyday experiences. For example, we know that a compass needle will always align itself in the same direction, parallel to the Earth’s magnetic field regardless of the distance it is from the Earth’s surface. Based on this, it would seem ridiculous to think that the orientation of two magnetic films would depend on their separation distance, however this is what has been observed in magnetic multilayered structures as the thickness of the non-magnetic spacer layer is increased.

Several theories have had success in describing the IEC of ferromagnetic layers in these MML systems. The most well known of these models is an extension of the Ruderman-Kittel-Kasuya-Yosida (RKKY) model developed independently in the 1950’s by M.A. Ruderman and C. Kittel [9], T. Kasuya [10], and K. Yosida [11] describing the effect of a magnetic impurity atom on the conduction electrons of a nonmagnetic host metal. Other models, treats the conduction electrons of the nonmagnetic layer as particles in a quantum well with spin dependent potential barriers at the magnetic and nonmagnetic interfaces. The observed oscillations are the result of the allowed standing wave solutions to the electronic density within the nonmagnetic layer. These models are able to yield analytical results due to their relative simplicity and they both relate the period of oscillation to the Fermi surface of the bulk spacer material in the limit of thick spacer layer[12], which is valid for spacers thicker than \(\sim 10\) monolayers (a monolayer is a layer one atom thick
and is denoted by ML).

Experimentally, IEC has been investigated primarily by probing the ferromagnetic layers. While these types of experiments allow one to make measurements such as the coupling orientation between two magnetic layers as a function of spacer thickness, they do not probe the mechanism of this coupling, the induced polarization within the non-magnetic layer, directly. The small amplitude of the induced electronic polarization due to rapid decay away from the magnetic layer, as well as the physical size of typical samples (spacer thicknesses typically of only a few hundred angstroms) makes measurements of the induced polarization of conduction electrons within the non-magnetic spacer layer very difficult. Most methods that make quantitative measurements of polarization within the spacer material are either measuring average polarization across the entire spacer, or probe nonmagnetic layers grown on a ferromagnetic substrate. These two methods don’t provide any information about the period of oscillation of the conduction electron polarization. The signal obtained with the first method, which can be done with X-ray magnetic circular dichroism (XMCD)[13], is dominated by the first few monolayers (ML) close to the magnetic-nonmagnetic interface. To make measurements of the period of oscillation requires a technique that measures polarization of conduction electrons locally within the spacer. Techniques such as Mössbauer spectroscopy[14], perturbed angular correlation[15], and nuclear orientation[16, 17] provide methods of making local measurements, however their limited sensitivity restricts them to measurements close the magnetic-nonmagnetic interface where induced fields are strongest. In order to probe the behaviour deep within the nonmagnetic layer effectively requires a technique that can make sensitive measurements of the local polarization. The technique we have used to do this is beta-detected nuclear magnetic resonance ($\beta$NMR).

$\beta$NMR is a technique very closely related to conventional nuclear magnetic resonance (NMR). They differ in that the $\beta$NMR signal is generated by utilizing the $\beta$-decay properties of radioactive nuclei ($\sim 10^8$) that have been implanted directly into the sample, whereas NMR requires measurements on a large number ($\sim 10^{18}$) of the sample’s own nuclei to generate a signal. $\beta$NMR conducted at TRIUMF’s ISAC (Isotope Separator and Accelerator) facility uses radioactive $^8\text{Li}$, a spin 2 nucleus with a mean lifetime of 1.21 s. In the $\beta$-decay of a polarized radioactive $^8\text{Li}$ nucleus, a high energy electron is emitted preferentially along the direction of its nuclear polarization during the decay process $^8\text{Li} \rightarrow ^8\text{Be} + e^- + \nu_e$. The $^8\text{Be}$ produced in this process
quickly decays into two alpha particles, and do not contaminate the sample. ISAC has the capacity to produce a highly polarized low energy beam of $^8$Li. Furthermore, the implantation energy can be adjusted in the range 0.1-30 keV, corresponding to mean implantation depths of between 2 and 200 nm from the sample surface. This ability to conduct NMR experiments at either the surface, or into the bulk region of thin film nanostructures is what distinguishes $\beta$NMR from conventional, as well as related (such as muon spin rotation/relaxation ($\mu$SR)) NMR techniques.

$\beta$NMR experiments are conducted by placing the sample in a spectrometer. A schematic of the setup used in $\beta$NMR experiments can be seen in Fig 1.1. The most basic requirements for a $\beta$NMR spectrometer is a set of beta detectors placed in front and behind the sample in this geometry. Though not required, the sample is usually situated in a large longitudinal magnetic field, $H_0$, produced by a superconducting solenoid. A small oscillating magnetic field, $H_1$ which lies in the plane perpendicular to the polarization direction is applied by a small coil located near the sample. In conventional NMR, the signal intensity scales as the square of the resonance/oscillation frequency, $\omega^2$. This is because both the nuclear polarization in the sample and the induced electromotive force (EMF) in the induction coils scale with $\omega$. Since the $^8$Li beam is highly polarized before being implanted in the sample, and the measurements are not made with induced EMF, the $\beta$NMR signal is independent of applied magnetic field and could, in fact, be carried out in the absence of any $H_0$, provided that polarization is not lost too quickly to spin lattice relaxation. However it will be shown that conducting $\beta$NMR experiments with a large longitudinal field does have certain advantages.

There are two observables in $\beta$NMR experiments related to the static and dynamic properties of the local magnetic environment. Resonant experiments are carried out by monitoring the polarization of a continuously implanted beam of $^8$Li while scanning through a range of frequencies of the oscillating perpendicular field ($H_1$ in Fig 1.1). Loss of polarization occurs when the frequency, $\omega$, matches the Larmor frequency of the $^8$Li nuclei in the sample. The dynamic magnetic properties of a sample can be probed by implanting a pulse of $^8$Li and monitoring the relaxation of polarization, in the absence of the perpendicular field, as a function of time. Relaxation of nuclear spins in a metal is usually dominated by scattering of conduction electrons at the Fermi surface off of the nuclear spins mediated by the hyperfine coupling between the electron and the nuclear spin [18]. Typical resonance and relaxation spectra can been seen in Figs 1.2 & 1.3.
Figure 1.1: Basic setup of a βNMR experiment. Radioactive ions enter through a hole in the backward detector and are implanted into sample which sits in a longitudinal magnetic field, $H_0$. The asymmetry between the backward and forward detector counts are monitored as a function of $H_1$ oscillating magnetic field frequency, $\omega$. 
Figure 1.2: Typical $\beta$NMR spectrum obtained from a resonance experiment taken at $T=290$ K and $H_0=4.1$ T in GaAs.
Figure 1.3: Typical $\beta$NMR spectrum obtained from a relaxation experiment taken at $T=290$ K and $H_0=4.1$ T in GaAs.
Chapter 1. Introduction

The purpose of the project that is presented in this thesis was to use the βNMR technique to measure the hyperfine field distribution induced in a nonmagnetic Ag(001) due to an adjacent magnetic Fe (001) film grown with molecular beam epitaxy (MBE) on a GaAs (001) substrate. The data collected was compared with models based on current theories, as well as with results of experiments conducted on Fe/Ag MML with the similar, but complimentary technique of low energy muon spin rotation (LE-μSR)[19].

We begin by first reviewing the relevant theory of induced hyperfine fields in Fe/Ag MML, and how we use this to generate a model for the resonant spectrum we would measure in an experiment in Chapter 1. Next the experimental setup that is used, including to production and polarization of the radioactive probe nuclei and the spectrometer used in making our βNMR measurements, as well as how the Fe/Ag MML samples were prepared is discussed in detail in Chapter 2. Finally the results of the βNMR resonance experiments conducted on these samples and the form of the hyperfine coupling that we extract from comparison of the measured spectra with our model is presented is given in Chapter 3.
Chapter 2

Theory

In this chapter we introduce the model which allows us to calculate the $\beta$NMR frequency spectrum observed in Fe/Ag MMLs. The model depends on the distributions of implanted $^8$Li and hyperfine fields within the Ag layer. Stopping distributions used in the model come from Monte-Carlo simulations of $^8$Li$^+$ implantation into the sample. The spatial distribution of hyperfine fields in the nonmagnetic layer come from predictions based on the theoretical calculations that have been done in these systems. Creating a model resonant lineshape of $\beta$NMR measurements in these systems requires understanding of the hyperfine coupling between the nonmagnetic Ag and ferromagnetic Fe layers, as well as the physics of nuclear magnetic resonance, and $\beta$NMR specifically. A review of these areas will be given first before discussing how we use them to generate a model of the $\beta$NMR frequency spectrum.

2.1 Induced Hyperfine Fields in Fe/Ag (001)

The oscillatory nature of the coupling between ferromagnetic layers separated by a non-magnetic layer has been theoretically examined by several different methods. Attempts have been made to calculate the coupling between magnetic layers by attributing the difference in the total energy of the system for ferromagnetic and antiferromagnetic orientations, either \emph{ab initio} [21] or using tight binding approximations [22]. Obtaining conclusive results in this manner is difficult because the energy difference between the orientations is typically several orders of magnitude smaller than the total energy itself and they also give very little understanding of the physical mechanism of the oscillatory nature of the coupling.

Other models that have been proposed as a possible mechanism for this coupling are based on quantum confinement of the conduction electrons in the nonmagnetic layer [12, 23]. In these models, spin dependent reflection amplitudes at the magnetic/non-magnetic interfaces, along with the finite thickness of the nonmagnetic layer lead to a situation analogous to a quantum
well. The quantum interference due to multiple reflections at the interfaces leads to modification of the electron density of states in the form of standing waves.

The mechanism to describe the oscillatory IEC in MML structures that has had the most success has its origin in the theory which describes the interaction of a magnetic impurity with the conduction electrons of the host metal in which it sits, known as the Ruderman, Kittel, Kasuya, and Yoshida (RKKY) interaction [9, 10, 11]. In the case of a magnetic impurity in a nonmagnetic host metal, hybridization between the s-p conduction electrons of the host metal and the d electrons of the magnetic impurities results in an effective exchange interaction at the site of the impurity. The typical coupling for s-p/d hybridization is antiferromagnetic, and the surrounding conduction electrons attempt to screen the impurity spin. The electrons close to the impurity over-screen the spin by aligning anti-parallel, the electrons further out align parallel to the impurity which over-compensates the over-screened region, so the electrons further still align anti-parallel again, and so on out radially to infinity with decreasing amplitude. In three dimensions, the amplitude of these oscillations decays away like \( r^{-3} \), where \( r \) is the radial distance away from the impurity.

The spherical shells of alternating polarization arise due to the cutoff in k values for an electron in a metal. The occupied k-vectors extend from zero to the Fermi wave vector, \( k_F \), in the Fourier k-space of the host metal. A delta function for localized screening in real space requires all k-vectors:

\[
\delta(r) \propto \int_{-\infty}^{+\infty} e^{ikr} dk
\]

However, only electrons with k-vectors up to \( k_F \) are available for screening which means the host conduction electrons cannot possibly screen the impurity spin perfectly to the atomic scale, resulting in oscillations of the electronic polarization [25]. The period of oscillation is determined by the Fermi surface of the of the host metal, which should be expected as these oscillations originate because of the sharp cut-off in occupied k-states at the Fermi energy. The period of the oscillation in the electron polarization, \( \lambda_F \), is given by \( \pi \) divided by the wave-vector(s) that define the Fermi surface of the host metal, \( k_F \). The theoretical coupling of an electron to a magnetic impurity can be seen in Fig 2.1.

Two impurities will interact with each other when they are close enough to allow appreciable overlap of the screening polarization regions. If the
Figure 2.1: The oscillatory coupling strength as a function of distance (measured in multiples the unitless quantity $k_F r$) between a host conduction electron and a magnetic impurity predicted by RKKY theory. Taken from [25]
second impurity is situated such that it is in the region where the induced polarization of the electrons is spin-down (with respect to the first impurity spin direction), it is favoured to orient itself opposite to the polarization directions, resulting in ferromagnetic coupling between the two impurities. Alternatively, the impurities will couple antiferromagnetically if one impurity finds itself in a region where the other has induced spin-up electronic polarization.

Extension of the RKKY theory of magnetic impurities to MML have shown that these systems share many characteristics with the theory describing magnetic impurities. However, these structures present a more complicated situation since the superposition of the interactions of conduction electrons with the atoms in the bulk of the magnetic layer as well as the interface is necessary to properly describe the behavior of the entire system. The interface atoms will have the largest effect on the interaction as these atoms are most strongly coupled to the conduction electrons of the spacer layer, and it is therefore reasonable to include only the first few magnetic layers in the theory [25]. Calculations carried out assuming a spherical Fermi surface (free electron approximation) have led to the same relationship between the period of oscillation and the Fermi surface of the spacer, but the amplitude of the oscillations decay as $r^{-2}$ away from the magnetic layer [24].

In real metals the Fermi surface has a much more complicated shape than the simple sphere predicted in this approximation. The characteristic k-vectors that define the period of oscillation are called the critical spanning vectors. Spanning vectors are defined as vectors parallel to the interface normal that connect two points on the Fermi surface, one point having a positive component of the velocity in the interface direction and the other a negative component. A critical spanning vector is a spanning vector that connects two points of the Fermi surface where its gradient is perpendicular to the interface, meaning that the critical spanning vector(s) connects parallel regions of the Fermi surface[27]. For the (001) direction of a face centered cubic (fcc) metal, such as Ag, there are two critical spanning vectors which span the “neck” and “belly” regions of the Fermi surface and give rise to long and short period oscillations, respectively. The Fermi surface and critical spanning vectors can be seen in Fig 2.2.

There was initially some confusion as to whether the oscillations were actually related to the Fermi surface, as the periods predicted by this theory were smaller than those measured in experiment [29]. This problem was resolved by considering an effect called aliasing. Aliasing arises from the
Figure 2.2: Fermi surface and spanning vectors of a fcc lattice. The Fermi surface in the first Brillouin Zone is shown on the right, with the ”necks” in the (111) region shown in grey. The repeated cross section (taken through the slice represented by the dashed rectangle) is shown on the left, with the Fermi surface shown by the dark curves. The reciprocal lattice vector in the (001) direction is given by the white arrow, while the critical spanning vectors, \( q_{\perp}^{i} \) spanning the ”belly” and \( q_{\perp}^{s} \) spanning the ”neck” regions are given by the grey arrows. Taken from [29]

Fact that the thickness of a sample is not continuous on an atomic scale. The spacer thickness must increase in unit thicknesses of half the lattice constant, as this is the thickness of a single atomic layer. Since the thickness will increase in atomic units, experiments effectively sample the oscillations at discrete intervals determined by the thickness of a single monolayer (ML), \( D_0 \). Aliasing gives rise to longer periods than predicted by simple theory as can be seen in Fig 2.4. The coupling period of two magnetic layers can be measured by growing a wedge of nonmagnetic metal on a ferromagnetic substrate, then covering the wedge with another ferromagnetic layer. The period of oscillation is determined by measuring the orientation of the magnetization across the top ferromagnetic layer using Scanning Electron Microscopy with Polarization Analysis (SEMPA), which will oscillate due to the increasing
Chapter 2. Theory

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spacer thickness of the wedge separating it from the bottom ferromagnetic layer. Results of an experiment of this kind can be seen in Fig 2.3. Taking aliasing into consideration brings theoretical and experimental periods into better agreement with each other [30, 31] and confirms that the oscillations really are determined by the Fermi surface of the nonmagnetic spacer.

When one takes aliasing into consideration the theoretically predicted periods using critical spanning vectors from de Haas-van Alphen results for Ag (2.38 and 5.58 ML, or 4.8 Å and 11.3 Å[26]) agree very well with experimentally determined periods measured with SEMPA are 2.37±0.007 and 5.73±0.05 ML (4.8±0.1 Å and 11.6±0.1 Å)[32] for Fe/Ag(001).

From this description, we would expect the hyperfine fields in the nonmagnetic layer away from the magnetic/nonmagnetic interface resulting from the induced electron polarization to be of the form:

\[ B_{hf}(x) = B_0 \sum \alpha_i \sin(2\pi x/\lambda_i + \phi_i) \] (2.1)

where \( x \) is the perpendicular distance into the Ag, \( \lambda_i \) are the oscillation wavelengths given above, \( C_i \) and \( \phi_i \) are the amplitude and phase of each oscillation, and \( \alpha_i \) are the power law exponents determining how the oscillations decay away from the interface. For fcc Ag, the sum would be carried out over the two periods determined from the critical spanning vectors (ie: \( i=1,2 \)). This form of the induced fields in Fe/Ag is shown in Fig 2.5.

Theoretical descriptions that predict coherent oscillations in the electronic polarization of the spacer layer are based on perfectly flat and sharp interfaces. Even with the sophisticated techniques for growing thin films, such as molecular beam epitaxy (MBE), ideally perfect samples with flat, sharp interfaces are impossible. A model of what we expect the distribution of magnetism fields in the Ag layer based on this ideal oscillatory form in an external field of \( B_{ext}=3 \) T and assuming a Gaussian \(^8\)Li stopping distribution is shown in Fig 2.6. It shows sharp peaks that correspond to the peaks and troughs of the oscillating hyperfine fields seen in Fig 2.5. The two major peaks on either side of the external field are due to the region far from the Ag/Fe interface where there is less damping between adjacent peaks/troughs, while the satellite peaks at higher/lower fields than the \( B_{ext} \) are due to peaks/troughs closer to the interface where the fields are larger in magnitude.

Imperfections at the interface due to steps, interdiffusion, and strain have been examined in the context of the RKKY model by several groups and all have found that deviations from ideal interfaces greatly reduce the oscillations
Figure 2.3: Shows the wedged structure used to measure IEC period in Fe/Au/Fe, as well as results for the period oscillation of the coupling between the magnetic layers obtained using Scanning Electron Microscopy with Polarization Analysis (SEMPA). Taken from [29].
Figure 2.4: Shows the effect discreet layer thickness, resulting in the measured period of oscillation being longer than theoretical predictions. The dotted line shows the short period oscillations predicted by RKKY, the squares shows the values taken at discreet thicknesses, and the longer period oscillations that would be observed is shown with the solid line. Taken from [29]
Figure 2.5: Schematic of oscillations in the induced hyperfine fields in Fe/Ag in an external magnetic field $B_{ext}$ based on the RKKY theory of magnetic impurities in a nonmagnetic host.
Figure 2.6: Model distribution of magnetic fields expected for oscillatory hyperfine fields in an external fields $B_{ext}=3$ T and assuming a Gaussian $^8\text{Li}$ stopping distribution. The sharp peaks at higher/lowers fields than $B_{ext}$ correspond to peaks/troughs in the induced hyperfine fields, with the two large, central peaks coming from the region far from the Ag/Fe interface where damping is smallest.
[12, 34, 37]. Roughness as a result of atomic terraces separated by steps has been shown to wipe out the short wavelength oscillations [38] since there would no longer be a well-defined $x$ in Eq. (2.1). Furthermore, a vertical mismatch between atomic planes, as little as 0.8% for Fe/Ag(001) [39], can lead to suppression of both the long and short wavelengths.

For reasons that will be discussed in section 3.7 of the next chapter on Sample Preparation, we feel that the Fe/Ag interface of the samples being studied is disordered enough that coherent oscillations will not be present over lateral distances we measure over. We predict that this will act to “smear” the fields between the $\pm x^{-\alpha}$ envelope.

The form of hyperfine field predicted by RKKY theory is divergent at the Fe/Ag interface, which is unphysical. Calculations of the induced hyperfine field in the Ag layer closest to the interface have been done [40] which give a value of $\sim 300$ kG for the hyperfine field at the site of the Ag nucleus closest to the interface. However, in general, Ag and Li will have couple differently to the hyperfine fields of the Fe. In our experiment, we would expect the fields to approach the hyperfine coupling of $^8$Li in Fe, which we expect to be on the order of a few kG, based on measurements of the induced hyperfine field at the site of a Li impurity in a substitutional site of Fe [41]. We have therefore chosen to use a form:

$$B_{hf}(x) = \frac{B_0}{1 + (x/\lambda_F)^\alpha}$$  \hspace{1cm} (2.2)

where $\lambda_F = 2\pi/k_F$ is taken as the long period Fermi wavelength of Ag (11.6 Å) makes $x/\lambda_F$ a unitless quantity. This form behaves like $x^{-\alpha}$ far from the Fe/Ag, but avoids the asymptotic behavior at small $x$. This form of hyperfine coupling can be seen in Fig 2.7.

### 2.2 Principles of $\beta$NMR

$\beta$NMR is a technique that exploits the phenomenon of nuclear magnetic resonance (NMR) to make measurements of local internal electronic and magnetic environments. NMR techniques can be employed on any particle with non-zero magnetic moment. The magnetic moment of a nucleus, $\vec{\mu}$, is directly proportional to its spin, $\vec{\mu} = \gamma \hbar \vec{I}$, where $\vec{I}$ is the spin of the nucleus, and $\gamma$ is known as the gyromagnetic ratio and is a proportionality constant specific to any nucleus. For $^8$Li $\gamma = 6301$ kHz/T. Simple classical electrodynamics tells
Figure 2.7: Schematic of the induced hyperfine fields with coherent oscillations suppressed by interface roughness in Fe/Ag resulting in a “smearing out” of the induced magnetism over the shaded area in the Ag.
us that in the presence of an external magnetic field, $\vec{H}$, $\vec{\mu}$ will experience a torque given by $\vec{\mu} \times \vec{H}$ [20]. Since torque is defined as the rate of change of angular momentum, which can be expressed in terms of magnetic moment, we have the equation of motion:

$$\frac{d\vec{\mu}}{dt} = \vec{\mu} \times \gamma \vec{H} \tag{2.3}$$

This implies that the instantaneous change in $\vec{\mu}$ is perpendicular to both $\vec{\mu}$ and $\vec{H}$. If $\vec{H}$ is a static field, $\vec{\mu}$ will sweep out a cone shape as it precesses around the magnetic field direction. The rate of this precession is known as the Larmor frequency, $\omega_L$, and is found by solving the equation of motion above. A convenient way to solve this equation is to consider a rotating frame of reference where $\vec{\mu}$ is at the origin, and $\vec{H}$ lies along the z-axis. If this reference frame rotates with instantaneous angular velocity $\vec{\Omega}$, then it can be shown that in this reference frame $d\vec{\mu}/dt$ can be written as [18]:

$$\frac{d\vec{\mu}}{dt} = \frac{\delta \vec{\mu}}{\delta t} + \vec{\Omega} \times \vec{\mu} \tag{2.4}$$

Making this change in Eq. (2.3) gives:

$$\frac{\delta \vec{\mu}}{\delta t} + \vec{\Omega} \times \vec{\mu} = \vec{\mu} \times \gamma \vec{H} \tag{2.5}$$

or

$$\frac{\delta \vec{\mu}}{\delta t} = \vec{\mu} \times (\gamma \vec{H} + \vec{\Omega}) \tag{2.6}$$

This equation of motion in the rotating frame is identical to the equation in the lab frame (non-rotating), except we now have an “effective field”:

$$\vec{H}_e = \vec{H} + \vec{\Omega} / \gamma \tag{2.7}$$

For a given external field, $\vec{H} = H_0 \hat{z}$, we can find $\omega_L$ of $\vec{\mu}$ by making $\Omega = \omega_L$. Since this means that our frame of reference and $\vec{\mu}$ are rotating at the same rate around $\vec{\mu}$, we have $\delta \mu / \delta t = 0$. From our equation of motion in this frame, this requires that $H_e = 0$, which implies that $\vec{\Omega} = -\gamma \vec{H} = -\gamma H_0 \hat{z}$. This tells us that the Larmor frequency must be $\omega_L = \gamma H_0$.

Now, if we applied a magnetic field, $H_1$, that rotates in the xy-plane of the laboratory frame at the Larmor frequency, in our rotating frame this will be a static field that the spin will begin to precess about, which results in an
oscillating nuclear polarization along the $\hat{z}$ direction in the lab frame. When there are many nuclei with random initial phase angles, then this precession is seen as a loss of $\hat{z}$ polarization, since we observe that average all nuclei.

One can arrive at this very same result by approaching this problem using quantum mechanics. When a magnetic moment is in the presence of a magnetic field, $\vec{H}$, it will experience a Zeeman interaction energy given by $-\mu \cdot \vec{H}$. The Hamiltonian that describes a nucleus in the presence of an external magnetic field is:

$$H = -\gamma \hbar H_0 I_z$$

(2.8)

where we take the external field to lie along the $z$-direction, and have magnitude $H_0$. We can see that the magnetic field lifts the degeneracy of the $I_z$ eigenvalues, $m_I$, which are the projections of $\vec{I}$ along the $z$-direction:

$$m_I = -I, -I + 1, ..., I$$

The energy levels are given by,

$$E = -\gamma \hbar H_0 m_I$$

(2.9)

This tells us that in the presence of a magnetic field a non-zero spin nucleus is in a lower energy state when it is aligned with that field. Quantum mechanics tells us that by introducing a time dependent perturbation, in the form of an alternating magnetic field perpendicular to the static field $\vec{H}$, transitions can occur between adjacent levels if the angular frequency of the perturbation is equal to $\gamma H_0$. This is exactly the value of the frequency we found for the rotating magnetic field that would result in loss of polarization along $\hat{z}$ using a classical description above.

In real materials, the local magnetic environment of the spin is a superposition of the external applied field with the internal magnetic field due to the surrounding nuclei and electrons. These internal magnetic fields shift the Larmor frequency from the frequency one would get with just the external magnetic field. In a non-magnetic metal the frequency shift is due to the Pauli paramagnetic susceptibility of the conduction electrons and is known as the Knight shift. The external magnetic field causes polarization in the electrons, which produces an internal magnetic field, $\Delta H$. $\Delta H$ scales with the strength of the applied field, $H$, however the ratio $\Delta H/H$ is a constant characteristic of the host metal [18]. In Fe/Ag, the presence of the Fe layer
will induce polarization of the conduction electrons in the Ag layer. Since electrons have a relatively large magnetic moment, this polarization leads to a distribution of internal magnetic fields within the normally nonmagnetic Ag which will lead to a distribution of Larmor frequencies of the nuclei in this region.

In conventional NMR, nuclear polarization is generated by the application of a large magnetic field. The distribution of $m_I$ spin projections is given by the Boltzmann distribution. The perpendicular magnetic field oscillating at frequency $\omega$ is applied using induction coils in the xy-plane. In typical NMR experiments the Larmor frequency is in the radio-frequency (RF) range, so it is common to refer to the perpendicular oscillating magnetic field as simply an RF field. After some time the RF field is shut off and for $\omega=\omega_L$, the coils will pick up the induced voltage caused by the precessing spins in an excited state as they relax back to equilibrium. Because the magnetic moment of an individual nucleus is small, a large number of spins ($\sim10^{18}$) are required to generate a signal. Since the polarized nuclei represent a small fraction of the total sample, conventional NMR is generally limited to studies of the bulk properties of large samples where many nuclei are available.

$\beta$NMR, while based on the same principle as NMR, is quite different from conventional NMR in several ways. Instead of measuring the resonance of polarized nuclei of the sample itself, we measure the resonance of implanted polarized radioactive nuclei which emit a high energy electron ($\beta$-particle) preferentially along their spin axis. Measuring resonance in this way greatly increases the signal to noise ratio which means that only $\sim10^8$ spins are required to generate a typical $\beta$NMR spectrum. This also means that the restrictions of conventional NMR on the size of a sample do not apply to samples in $\beta$NMR experiments. It is possible to implant $^8$Li in samples with thicknesses from as thin as a few tens of angstroms grown on substrates less than 2-3 mm, and with lateral dimensions larger than the $^8$Li beam spot, which has a diameter of $\sim4$ mm. Also, since the implanted nuclei have a high degree of polarization (60%), the large magnetic field characteristic of conventional NMR is unnecessary, which means it is possible to measure only the internal magnetism of a sample.

In $\beta$NMR experiments, we apply a continuous wave (CW) RF at one frequency while constantly implanting $^8$Li into the sample. Time integrated counts ($N_{F/B}$) are made in the front (F) and back (B) $\beta$ detectors shown in Fig 1.1 for a period of time (on the order of several $^8$Li lifetimes), then the RF is stepped to the next frequency and this procedure is repeated throughout
the range of frequencies we are interested in. The counts in each detector for a particular RF, $N_{F/B}$ is the time averaged integral of the $\hat{z}$ component of the $^{8}$Li polarization weighted by the decay of the radioactive $^{8}$Li. We observe the time averaged polarization since the $\beta$-particle is emitted along the spin direction of the radioactive nuclei at the time it decays. The form of this is given by [35]:

$$N_{F/B} = N_{F/B}^0 \int_0^\infty \frac{1}{\tau} e^{-\frac{t}{\tau}} [1 + A_{F/B} G_{zz}(t)] dt \quad (2.10)$$

where $N_{F/B}^0$ is determined by the intensity and polarization of the $^{8}$Li beam as well detector geometry, $\tau$ is the $^{8}$Li lifetime, $A_{F/B}$ is the asymmetry parameter for the particular detector/sample geometry, and $G_{zz}(t)$ is the time dependent polarization along the $\hat{z}$ direction for $^{8}$Li in a static magnetic field also directed along $\hat{z}$. The function $G_{zz}(t)$ is a function of the populations and spin dynamics at the $^{8}$Li sites, as well as transitions and any other interaction that would affect the polarization over time.

The polarization of the implanted $^{8}$Li is monitored the asymmetry between the front and back detectors at a particular RF frequency by combining the counts in the two detectors in the following way:

$$A = \frac{N_F - N_B}{N_F + N_B}. \quad (2.11)$$

Off resonance, the majority of $^{8}$Li will be polarized in the $+\hat{z}$ direction, resulting in more counts in the front detector, however, on resonance we expect a loss in the asymmetry since the $^{8}$Li is just as likely to be pointing in the $-\hat{z}$ direction as the $+\hat{z}$ direction when it decays. Ideally the off resonance asymmetry, called $A_0$, would take the values of $\pm 1$, depending on the polarization direction of the implanted $^{8}$Li, since all the counts are in either the front or back detectors. When the RF is on resonance we expect the time averaged polarization along the $\hat{z}$ to be zero resulting in a loss of asymmetry. In practice $A$ is not $\pm 1$ on resonance, and zero off as it is a function of the geometry of the detectors as well as the polarization and nuclear decay properties of the $^{8}$Li.

In the absence of any quadrupolar splitting, such is the case for Ag, the form of the $z$-component of the polarization, $P_z$, can be determined similar to the polarization of implanted muons in $\mu$SR which can be found in Refs [35, 36]. To do this we take a reference frame that is rotating about the $\hat{z}$ axis with the same angular frequency as the applied RF, $\omega$. The effective
Chapter 2. Theory

field, $\vec{H}_{\text{eff}}$, that results from this is the modified $\hat{z}$ component from Eq. (2.7), as well as the RF field $\vec{H}_1$ in the $\hat{x}_R, \hat{y}_R$ plane, where $(\hat{x}_R, \hat{y}_R, \hat{z})$ are the unit vectors of the rotated reference frame:

$$\vec{H}_{\text{eff}} = \frac{\omega_L - \omega}{\gamma_L} \hat{z} + H_1 \dot{\rho}(\phi)$$

where $\omega_0 = \gamma_L H_0$ is the Larmor frequency and $\dot{\rho} = \hat{x}_R \cos \phi + \hat{y}_R \sin \phi$. $\phi$ is the angle between $\vec{H}_1$ and the x-axis in the rotating frame, $\hat{x}_R$. With $H_{\text{eff}}$ directed along $\hat{r} = \hat{z} \cos \theta + \hat{\rho} \sin \theta$, and assuming initial polarization along the $\hat{z}$ direction, the time evolution of $P_z$ as a function of $\omega$ is:

$$P_z = \cos^2 \theta + \sin^2 \theta \cos(\omega_{\text{eff}} t)$$

where $\omega_{\text{eff}} = \sqrt{(\omega_0 - \omega)^2 + \omega_1^2}$ is the precession frequency in the effective field, $\omega_1 = \gamma_L H_1$ is the precession in the RF field and $\cot \theta = (\omega_0 - \omega)/\omega_1$.

The time dependence of $P_z$ is in the $\cos(\omega_{\text{eff}} t)$ term, so the time averaged polarization will be the form of the Laplace transform of $\cos$:

$$\int_0^\infty e^{-st} \cos at = \frac{s}{s^2 + a^2}$$

This form is what will be found for the asymmetry in the F/B detectors under RF excitation by taking Eq. (2.10) with $P_z(t)$ incorporated into the function $G_{zz}(t)$. Applying this to Eq. (2.11), one obtains a Lorentzian lineshape for the RF induced asymmetry:

$$A_{RF} = A_{RF}(0) \frac{\Delta^2}{(\omega_0 - \omega)^2 + \Delta^2}$$

where $A_{RF}$ is the peak amplitude, and $\Delta^2 = \frac{1}{\tau^2} + \omega_1^2$ is the half width at half maximum.

### 2.3 Modeling the $\beta$NMR lineshape in Fe/Ag (001)

Given the internal magnetic field distribution in the Ag layer as a function of distance from the Fe/Ag interface, as well as the $^8$$\text{Li}$ stopping distribution in the Ag layer, one can generate a phenomenological $\beta$NMR lineshape that one might expect to measure in an experiment. One can approximate the
distribution of $^8\text{Li}$ stopping in the Ag layer with a Gaussian distribution because the profiles of $^8\text{Li}$ calculated by Monte-Carlo software, TRIM.SP, fits extremely well to this form. The model lineshape based on this information can be used in a fitting routine to find the induced field parameters that most closely replicates the experimental data.

The model lineshape we want to make is a distribution of the number of $^8\text{Li}$ that will stop in the presence of internal magnetic field $B$, $n(B)$. For static (time-independent) internal fields, the resonance lineshape is closely related to the internal magnetic field distribution, $n(B)$. In a $\beta$NMR resonance experiment, the $^8\text{Li}$ polarization is monitored while applying a perpendicular magnetic field at frequency $\omega$, as explained in the previous section. As was shown, the resonant frequency of a nucleus is proportional to the magnitude of the local magnetic field. This means that the $n(B)$ of our model should be proportional to the $n(\omega)$ measured in the experiment.

The magnetic fields in the Ag layer that the implanted $^8\text{Li}$ experience is the induced hyperfine field, $B_{hf}$, and the longitudinal field of $B_{ext} \sim 4.1$ Tesla (T) applied normal to the surface of the sample. It has been found by Low-Temperature Nuclear Orientation (LTNO) [16, 17] that in the presence of such a magnetic field directed out of the surface plane, the Fe magnetization as well as the induced hyperfine fields in the Ag layer, are also directed out of the plane. The total internal magnetic field will be the induced positive and negative hyperfine fields centered on the large external field.

$$B(x) = B_{ext} + B_{hf} \tag{2.15}$$

The “smearing” effect of $B_{hf}$ due to sample imperfections means that the magnetic field at a given $x$ can lie anywhere between the maximum and minimum fields given by:

$$B_{\text{max}}(x) = B_{ext} + \frac{B_O}{1 + (x/\lambda_F)^\alpha}; \tag{2.16}$$

$$B_{\text{min}}(x) = B_{ext} - \frac{B_O}{1 + (x/\lambda_F)^\alpha}$$

This distribution of fields is shown in Fig 2.7. We isolate $x$ in this equation to obtain:

$$x(B) = \lambda_F \left[ \left( \frac{B - B_{ext}}{B_O} \right) ^\alpha - 1 \right] \tag{2.17}$$
Figure 2.8: \( x_i \) is the upper limit of the distance from the Fe/Ag interface where the internal field could have the value \( B_i \). By integrating the stopping distribution from \( x=0 \) to \( x=x_i \) gives the number of \(^{8}\text{Li} \) that could stop in a region of magnetic field \( B_i \).
Figure 2.9: Integration of the stopping profile from $x=0$ to $x=x_i$ gives the relative number of $^8\text{Li}$ that could stop in a region with a magnetic field of $B_i$. 
This function tells us that all the $^8$Li which stops between the Fe/Ag interface ($x=0$) and $x(B)$ could be at a site where the magnetic field is a particular value of $B$. Fig 2.8 shows this schematically. The relative number of $^8$Li that lie in this range can be found by integrating the normalized Gaussian of the stopping distribution, see Fig 2.9:

$$n(B_i) = \int_{0}^{x_i} e^{-\frac{x'^2}{\Gamma^2}} \, dx'$$

$x_c$ and $\Gamma$ are the center and the full width-half maximum of the stopping distribution, and are determined by simulating implantation of $^8$Li at a given energy. Repeating this through a range of $B$ values centered around the applied magnetic field $B_{\text{ext}}$ will generate the model lineshape. This can be seen in Fig 2.10. In $\beta$NMR experiments we measure a loss of polarization along the $\hat{z}$ direction, we expect maximum signal at frequencies which relates to where $n(B)$ is small, since few $^8$Li will be in resonance here. Therefore when we compare experimental results to this model, we construct the function

$$A = A_0[1 - a \times n(\omega)]$$

The baseline, $A_0$, is the off resonance polarization, and $a$ is a constant that relates the amplitudes of the model and the experimental signals.

One question that arises is whether this “static field” picture is valid, or if spin-lattice relaxation will skew the results by “magnifying” slower relaxing lines, such as in the Ag layer far from the Fe, over faster relaxing lines, such as in and near the Fe layer. Preliminary results from $\beta$NMR relaxation experiments suggest that, while spin-lattice relaxation at room temperature is approximately twice as fast in the Ag of these magnetic samples ($1/T_1 \sim 0.6$ s$^{-1}$) compared to a similar Ag film grown on a nonmagnetic substrate ($1/T_1 \sim 0.3$ s$^{-1}$), this relaxation is slow enough that this “static field” approximation is valid.
Figure 2.10: By taking many values for $B$, centered around $B_{\text{ext}}$ generates the distribution $n(B)$, which is our model of the $\beta$NMR lineshape expected from theoretical hyperfine field distribution and stopping distribution.
Chapter 3

Experiment

The technique of βNMR is distinct from conventional NMR in that polarized radioactive ions are implanted directly into the sample and used as the probe rather than the host nuclear spins. A radioactive ion beam (RIB) of polarized $^8$Li is the NMR probe used in βNMR experiments at TRIUMF’s ISAC facility. The RIB is produced using, what is called, the isotope separation on-line (ISOL) technique. The ISOL system, comprised of a primary production beam, a target/ion source, mass separator, and a separated beam transport and acceleration system, is capable of producing very intense ($\geq 10^8 \, \text{s}^{-1}$) RIB of $^8$Li suitable for βNMR experiments [42, 43].

3.1 $^8$Li$^+$ Production

ISAC experimental hall, where βNMR experiments are conducted, is housed in a separate building from TRIUMF’s cyclotron hall. A proton beam line (BL2A) carries the high-energy protons from the cyclotron to one of the two target stations housed in the ISAC building. This proton beam is what drives the ion production in the ISOL method. BL2A is capable of delivering a 100 $\mu$A proton beam extracted from TRIUMF’s 500 MeV H$^-$ cyclotron. The ISOL target is composed of a material of large atomic mass such as silicon-carbide (SiC) or Tantalum (Ta). Many different isotopes are produced through nucleosynthesis when the target is bombarded with high energy protons. Typically the high energy protons cause fragmentation of the nuclei within the target. The target is heated uniformly to high temperatures (~2000 °C) to enhance diffusion to the target’s transfer tube (see Fig 3.1 in a time comparable to the radioactive lifetime of the nuclei created [44, 45]. The target temperature is high enough to efficiently release the exotic nuclei, but not so high that the target material itself is desorbed through evaporation. The free radioactive nuclides are ionized by coming into contact with a heated tungsten extraction tube. The target is held at high voltage so that the extracted ions leave the target at an energy of 30 keV, typically,
and with a very small energy spread (1-2 eV), which is an important feature of the surface ionization sources used at ISAC. Having an ion beam with a narrow energy distribution greatly increases the efficiency for transportation and generating nuclear polarization. The entire target assembly can be seen in Fig 3.2.

The large operational and residual radiation fields produced by proton bombardment on the target material requires that the target stations be housed in a shielded target hall that is isolated from the experimental areas. All highly activated and potentially contaminated components, including production targets, ion sources, and beam dump as well as the decontamination and storage facility are located in the target hall. ISAC uses remote handling of the target modules to allow quick access to the production target which have high levels of residual activity and could be contaminated with.
Figure 3.2: Figure of ISAC surface ion source. The target and transfer tube in Fig 3.1 can be seen in the center left part of the diagram, with the system that extracts the ions shown to the right. Cooling lines prevent components from overheating. [49].
Figure 3.3: Figure of removable target module used in ISAC’s ISOL method for ion production. The surface ion source, Fig 3.2 is housed in the bottom most part of the module. The module is designed to be handled remotely, allowing targets to be changed with very little downtime. [48]
mobile activity. The target hall was also designed with two target stations, allowing the proton beam to be sent from one target station to the other, which greatly reduces the amount of time the RIB is off when targets need to be changed or maintenance on the target station is required. The target module is made of a 2 m long shielding plug on the bottom of which is mounted the target, ion source and extraction system, as shown in Fig 3.2 [47], see Fig 3.3.

In order to extract a beam of pure $^8\text{Li}$ from the large number of different isotopes produced at the target, all the ions from the ion source must be passed through a mass separator. The layout of the mass separator used at ISAC can been seen in Fig 3.4.

The mass separator consists of three main sections. The first section of the mass separator transports the beam from the target station to the mass separator. Electrostatic quadrupoles focuse the beam horizontally at HF1 and vertically in the center of B1. The second section, between HF1 and HF2, serves as a coarse mass separator. This section consists of a 60 degree bending element that accepts beam entering from either the east or west target. This bender is basically a region with a constant magnetic field oriented vertically that bends the path of the ions. An ion moving through this region of static transverse magnetic field will experience a Lorentz force perpendicular to both the ions velocity and the magnetic field, resulting in circular motion in the plane normal to the vertical magnetic field direction. Since the radius of curvature of the ion’s circular path is proportional the charge/mass ratio, the beam emerging from the bender will have dispersion. If all particles of the beam are singly ionized, then this dispersion separates the particles of different mass. Inserting slits into the beam at this point allows only the ions within a desired range of dispersion through.

The final section, lying between HF2 and HF4, is a high-resolution mass separator. By passing the beam through 2 pairs of identical 60 degree bending magnets high-resolution separation of masses is achieved. The sections between HF3 and HF4 and between HF2 and HF3 (Fig 3.4) are identical except that they bend the beam in opposite directions causing the dispersion of each to add. The dispersion of this last section is 5.2 cm per percent $\Delta M/M$, and the total dispersion of the entire mass separator is 5.8 and 4.6 cm per percent $\Delta M/M$ for the East and West targets, respectively. Here $M$ is the mass of the desired particle to be separated. This mass separator is designed to have mass resolution of up to 10,000 (mass resolution is defined as $M/\Delta M$, where $M$ is that mass of interest) [50]. Such mass resolution is more than is
Figure 3.4: Figure of the layout of the mass separator at ISAC. [50]
Figure 3.5: ISAC experimental hall, showing the Target Hall, and mass spectrometer underground, and the main experimental area where the $\beta$NMR area is situated.
required for separating light ions such as $^8\text{Li}$ (M here would be 8 atomic mass units(amu)), but does become necessary for separation of a specific isomer with much larger atomic weight that are used in other experiments carried out at ISAC.

The $^8\text{Li}$ beam exiting the mass separator is transported into the experimental hall where it is first polarized and then delivered to the $\beta\text{NMR}$ spectrometer. The target hall, mass separator and experimental area at ISAC can be seen in Fig 3.5.

### 3.2 Polarizer

The layout of the polarizer and the $\beta\text{NMR}$ spectrometers are shown in Fig 3.6. After passing through the mass separator the nearly monoenergetic (30 keV) RIB of $^8\text{Li}^+$ is electrostatically guided to the $\beta\text{NMR}$ beamline in the low-energy experimental area. Before the RIB can be used in $\beta\text{NMR}$ experiments, nuclear polarization must be generated in the “optical pumping region” section (see Fig 3.6) of the polarizer. The Na neutralization and He re-ionization cells, make up what is called the polarizer, which is shown in detail in Fig 3.7.

Nuclear polarization in the $^8\text{Li}$ RIB is achieved by optically pumping of the total spin states (F=5/2, 3/2) in neutral paramagnetic $^8\text{Li}$ with a beam of circularly polarized laser light collinear with the ion beam. Since the incoming beam is comprised of $^8\text{Li}^+$ ions, the first step in this process is to neutralize the beam by passing it through a Na vapour cell. $^8\text{Li}^+$ has a large cross section for charge exchange with Na ($\geq 10^{-15}$ cm$^2$) so by passing the beam through this cell we are able to achieve up to 90% neutralization with very little change in the beam emittance of $8\pi$ mm mrad, however neutralization under typical experimental conditions is more like 50-70%. The Na cell, which confines the Na vapour, is kept at a temperature of $\sim 450$ °C. This is the temperature that maximizes neutralization of the beam, see Fig 3.8. Upon exiting the Na cell the neutral beam then enters the drift region where the neutral $^8\text{Li}$ is polarized by optical pumping, and remaining ions are removed by charged plates [51, 52].

The neutral beam travels for 1.7 m through a drift region where 6 Helmholz coils keep a constant magnetic field of 1 mT collinear with the beam, which defines the axis of polarization. It is in this region that atomic excitation of the $^8\text{Li}$ by a counter-propagating circularly polarized laser beam produces
Figure 3.6: $\beta$NMR beamline at ISAC. An unpolarized $^8$Li beam enters on the far left then passes through the polarizer before being sent to one of the three experimental areas: the low or high field $\beta$NMR spectrometers, or the Osaka University experiment.
Figure 3.7: Polarizer section of the $\beta$NMR beamline where unpolarized $^8\text{Li}^+$ enters from the left and is neutralized in the Na vapour cell. Optical pumping by the laser achieves nuclear polarization in the beam which is then re-ionized in the He gas re-ionizer before exiting to the right.
Figure 3.8: Graph of $^7$Li$^+$ current vs. Na vapour temperature. The loss of current in this figure ($\sim 90\%$) is due entirely to neutralization of the stable $^7$Li ions. [51]
Figure 3.9: D1 atomic transition of $^8$Li that is optically pumped in the polarizer. Absorption of a circularly polarized photons always adds one unit of angular momentum ($\Delta m_F=1$), while relaxation results can either add or subtract one unit, or have no change ($\Delta m_F=0, \pm 1$). Many cycles will result in atoms being in the highly polarized $F=5/2$, $m_F=+5/2$ state.
nuclear polarization. The laser is tuned such that it optically excites the D1 atomic transition, \(2s^2S_{1/2} \rightarrow 2p^2P_{1/2}\) in \(^8\text{Li}\), which occurs at \(\lambda = 671\ nm\). A schematic of this electronic transition is shown in Fig 3.9.

Spin-orbit coupling between the nucleus and the valence electron in the neutral \(^8\text{Li}\) atom breaks the degeneracy of the total spin states, \(F=5/2\) and \(3/2\). The high polarization of the \(^8\text{Li}\) beam is achieved by pumping both of these hyperfine levels by two laser lines separated by 382 MHz, which corresponds to the energy difference between the two total spin states of the valence electron. Excitation by absorption of a circularly polarized photon with positive helicity requires \(\Delta m_F = +1\) so that overall angular momentum is conserved (circularly polarized photons are in state \(S=1, m=1\)). The atom will decay back to the ground state by spontaneous emission of a photon, obeying the selection rule \(\Delta m_F = 0, \pm 1\) since the emitted photon may have any allowed spin projection. Total spin polarization, and therefore nuclear polarization, is thus increased as the number of optical cycles increases. The short lifetime of the excited electron state (27 ns) compared to the transit time (2 ms) means that many optical cycles occur as the neutral \(^8\text{Li}\) drifts through this region \([53]\). Therefore, the beam that reaches the He cell is in the highly polarized total spin state \((F=5/2, m_F=+5/2)\) corresponding to the \(m_I=+2\) nuclear spin state, which is directed opposite to the propagation direction of the beam itself. The beam can also be polarized in the opposite direction in exactly the same manner using laser light of the opposite helicity. The beam polarization can be switched very quickly between the two helicities by inserting a quarter wave plate into the laser optics system.

The laser used for optical pumping during these experiments was a Spectra-Physics 3900S standing wave Ti:sapphire (TiS) laser pumped by an 18 W argon ion laser, though this system has since been updated by replacing the TiS laser with a dye laser. Lasing at \(\lambda = 673\ nm\) is accomplished by fitting the laser cavity with a rear mirror optimized for this wavelength and an \(R=99\%\) output coupler. The cavity length is designed so that longitudinal cavity modes are separated by 382 MHz and a 1mm thick, \(R=10\%\) intra-cavity etalon restricts operation to just the two modes. The resultant output is two laser lines required to pump the two electronic states, each with a power of \(\sim 100\ mW\). These two modes have instantaneous line widths of \(\sim 1\ MHz\) which are broadened to \(\pm 20\ MHz\) on the timescale of 1 s due to acoustic noise, which is still much narrower than the \(\sim 100\ MHz\) absorption bandwidth of the incoming beam at 30keV. This absorption bandwidth is due the energy spread of the neutral \(^8\text{Li}\) beam, typically a few eV as a result of the
inherent spread of the ion source (\(\sim 2\) eV) as well as collisions with Na atoms in the neutralization cell [51, 53]. \(^{8}\)Li atoms that pass through the polarizer at slightly different energies will “see”, in each atom’s frame of reference, photons of slightly higher/(lower) frequency, and therefore slightly higher/(lower) energy due to what is known as the Doppler effect. The Doppler width has the form:

\[
\delta \nu = \nu_o \frac{\delta E}{\sqrt{2 E m_c c^2}}
\]

For \(^{8}\)Li the resonant frequency is \(\nu_o = 4.47 \times 10^{14}\) Hz, and for the beam at ISAC, with energy \(E\) (30 keV) and width \(\delta E\) (2 eV), \(\delta \nu\) has a value of about 42 MHz [46]. Notice that this width is proportional to \(E^{-1/2}\), which, for thermal energies, would be enormous. Thus, the beam is accelerated to 30 keV to narrow the Doppler width, allowing efficient optical polarization of the beam to become feasible. The power of each laser line must be spread over this absorption width on a ms timescale, this being the time scale for \(^{8}\)Li to pass through optical pumping section of the polarizer. Resonant electro-optical modulators (EOM) of 19 MHz and 28 MHz in series are used to produce laser sidebands spaced at \(\sim 10\) MHz intervals, effectively broadening the linewidth of the laser. This spacing was found to pump the beam the most efficiently, with closer spacing only resulting in a minor increase in polarization. At Na temperatures of \(\sim 430\) °C (high vapour density, and therefore more broadening), the EOMs gives almost 50% increase in polarization, and nearly 70% at 370 °C [53].

To efficiently polarize the beam using this method it is crucial that the laser frequency remains extremely stable. Even slight changes in laser frequency can result in total loss of polarization. Drifts are prevented by using a feedback system with a 2 GHz free-spectral-range spectrum analyzer to monitor and maintain the frequency difference between the one of the TiS modes and a single-mode frequency stabilized He-Ne laser reference line. Any change in frequency will automatically be corrected by adjusting the cavity length by means of a piezo-mounted rear TiS mirror. Frequency stability is \(\pm 10\) MHz on average. This feedback system also monitors the number of TiS modes, and any extraneous modes are corrected for by adjustments to the intra-cavity etalon [51]. After the laser has been tuned and stabilized, fine-tuning of the polarization is done by slightly decelerating the beam by applying a bias voltage to the neutralization cell. Fig 3.10 shows \(\beta\)-decay asymmetry versus Na cell voltage, found by integrated \(\beta\) count measure-
ments for 10 s at each voltage. The three peaks seen are due to scanning 2 hyperfine lines across 2 equally spaced laser modes, with the central peak having the largest asymmetry amplitude corresponding to pumping of both hyperfine levels [51].

The final section of the polarizer is a He gas cell that re-ionizes the polarized neutral beam to allow the $^{8}\text{Li}$ to be steered and focused electrostatically onto the sample. Because the ionization cross-section of $^{8}\text{Li}$ with He is more than an order of magnitude smaller than for neutralization by Na the beam emerges with a larger emittance from the He cell, and means elements downstream must have a larger acceptance accordingly. Fig 3.11 shows scattering effect of the beam for no flow and 3 times the flow of He required for optimum ionization. It can be seen that He flow has very little effect on the center of the beam profiles, however the transverse momentum profile is significantly broadened from passing through the He cell [52].

An electrostatic bender placed after the He cell redirects the ionized beam by 45° towards another bender that then directs it to one of three experimental areas (the high and low field $\beta$NMR spectrometers, and the Osaka experiment, see Fig 3.6), while the undeflected neutral beam passes straight through to a neutral beam monitor. The ion flux that reaches the sample is $\sim 10^{7}$ s$^{-1}$.

### 3.3 $\beta$NMR High-Field Spectrometer

After being polarized and re-ionized, the beam can be sent to the spectrometer where it is used for $\beta$NMR experiments. The $\beta$NMR spectrometer, shown in Fig 3.12, is where the actual $\beta$NMR experiment takes place. The now polarized $^{8}\text{Li}^{+}$ beam enters from the left, passing through a hole cut out of the back detector before entering the last Einzel lens at the entrance of a high homogeneity 9 T superconducting solenoid. The profile of the beam on the sample, or “beam spot”, is extremely sensitive to the beam energy as well as the voltage on the Einzel lens, and the magnetic field within the solenoid. The beam spot is monitored using a CCD camera and imaging a piece of scintillating plastic located at the sample position. More thorough discussion of beam optics and beam spot will be given later on, but elements are tuned such that the beam spot is 3-4mm in diameter at the sample.

The $^{8}\text{Li}$ ions are implanted into the sample which is positioned in the center of the solenoid. Sample dimensions are typically 8mm by 10mm and
Figure 3.10: Asymmetry at 380°C for each laser helicity as a function of the voltage on the Na cell deceleration plates. The three peaks are due to scanning two hyperfine lines across two equally spaced laser modes, with the central peak corresponding to pumping both hyperfine levels. The drifting offset is due to the beam shifting position on foil.[51]
Figure 3.11: Measured emittance from He cell extrapolated back to the center. Shows no He flow on left, and He flow of 0.029 Torr litre/s on right. 10 contours are equally spaced from 5% to 95% of maximum phase space density[52]
Figure 3.12: βNMR high field spectrometer. $^8$Li ions enter from the left, through a hole in the backward detector, and are implanted into the sample situated at the center of 9 T superconducting solenoid.
limited to a thickness of a few mm by the necessity of beta particles, emitted in the radioactive decay of $^8$Li, to pass out through the sample and make it to the detector. Thermal contact with a He cold finger cryostat with an operational temperature range of 2-500 K allows the temperature control of the sample. Since many of the experiments carried out with this spectrometer are at cryogenic temperatures, the sample must be kept in an ultra high vacuum (UHV) environment. Otherwise residual gases would condense and build up on the sample surface. This would be enough to prevent nearly all incident $^8$Li from stopping within the sample at low energies. Also, the beam is not energetic enough to pass through a thin window, therefore the entire final leg of the beam line must also be UHV compatible. Differential pumping with large cryopumps reduces the pressure of $10^{-7}$ Torr upstream of the spectrometer, to the order of $10^{-10}$ Torr in the main UHV chamber.

The cryostat is mounted on a large bellows with a small motor so that it may be withdrawn from the magnet bore in order to change the sample through a load lock located on the top of the main vacuum chamber. This obviates the need to vent the entire vacuum chamber to atmospheric pressure during sample changes.

The spectrometer is designed longitudinally, such that the beam axis, $^8$Li polarization, and magnetic fields are all coaxial. A transverse field would act to beam the beam away from the sample, while the large longitudinal magnetic fields, several Tesla, used in the high field spectrometer act to focus the ions on the sample. Incoming ions and outgoing beta particles are strongly confined along the beam axis by the large magnetic fields in the spectrometer when the magnetic solenoid is on. The $\beta$-detectors are made of Bicron BC412 plastic scintillator and are approximately 0.6 cm thick. The front detector (located behind the sample, with respect to the incoming ions) is approximately 4 cm in diameter and is centered on the beam/magnetic field axis a few cm downstream of the sample. Confinement of the emitted betas inside the magnet bore means that the backward detector, approximately 20 cm by 14 cm by 0.6 cm thick must be positioned further upstream, outside of the solenoid, to ensure that the outgoing betas will be detected while still allowing the incoming beam to pass through a 1.8 cm slot cut out of the scintillating plastic. Each detector and its UVT plastic light guide is held in stainless steel housing with thin stainless steel windows that allow transmission of the high energy betas while isolating the detectors from the UHV chamber which allows them to be easily during bake outs. Light guides transport the photons, produced when a beta particle strikes the scintillating
plastic, to the photomultiplier tubes. The photomultiplier tubes convert the light from the scintillation to an electric pulse that is sent to the electronic counters. In this geometry, the detectors will subtend very different solid angles in the absence of magnetic field produced by the solenoid, however their efficiencies are very similar under experimental conditions due to the focusing effect of the solenoid.

The high field $\beta$NMR spectrometer has the ability to vary the implantation energy of the beam anywhere from 100 eV to 30 keV, which in turn, varies the mean stopping depth of $^8$Li from just a few nm from the sample’s surface to as deep as 200 nm into the bulk. This feature makes $\beta$NMR a very useful tool for studies of thin film nanostructures. Energy of implantation is adjusted by placing the spectrometer, magnet, cryostat and all other electronics on a ground insulated, high voltage platform which creates an electrostatic potential step up to 30 keV that the $^8$Li$^+$ ion must “climb” before implanting itself into the sample.

In Fig 3.12 everything located to right the “HV Isolator” is always at ground potential, while everything to the left lies on the high voltage platform and will be at high voltage when the platform is biased. The beam passes through a grounded tube surrounding the last Einzel lens and extends into the magnet bore. Deceleration of the beam occurs in the small space between this gold plated Einzel lens and the sample. Fig 3.13 shows an electric potential map of the region between the grounded tube (light grey) and the sample at high voltage (dark grey), and Fig 3.14 shows the potential step along the beam axis ($r=0$) in this region for an applied bias of 30 kV.

This allows the energy of implantation of the beam to be selected so that the majority of $^8$Li$^+$ stops at different distances from the magnetic interface in these samples, to build up a profile of the magnetic environment within a sample.

It has been shown that $^8$Li$^+$ implanted into fcc Ag will come to rest in one of two possible sites, the substitutional (S) and octahedral (O) sites, see Fig 3.15. At low temperatures, most of the $^8$Li stops in the O site, but as the sample is warmed to room temperature the $^8$Li$^+$ ions occupy mostly the S site, as can be seen in Fig 3.16, showing $\beta$NMR spectra taken in a 500 Å Ag film grown on SrTiO$_3$ [55, 56].
Figure 3.13: Electric potential map of the deceleration region of the spectrometer for platform bias of 30 kV. Dark grey shows the region that is at 30 kV, while light grey shows the region that is at ground (0 kV).[54]
Figure 3.14: Electric potential step along the beam axis (r=0) in the deceleration region of the spectrometer when platform is biased to 30keV.[54]
Figure 3.15: FCC lattice of Ag with three possible sites for $^8$Li to stop in; substitutional (S), octahedral (O), and tetragonal (T).[55]
Figure 3.16: $\beta$NMR spectra taken at temperatures between 15 K and room temperature showing the shift from $^8$Li ions stopping at the octahedral site at low temperatures to the substitutional site at high temperatures in thin Ag film grown on a MgO substrate.[55]
3.4 Beam optics and Beam Spot

Ideally, one would like a well-collimated beam with as small a beam spot as possible for $\beta$NMR experiments. To achieve this the beam must be focused on the sample by tuning the various focusing elements upstream. It was shown in Fig 3.11 that emittance of the beam is larger upon exiting the He gas cell. Before reaching the sample it is focused by three Einzel lenses and three adjustable collimators. These beam optical elements are located in the final section of the beam line between, the two 45° benders and the spectrometer. All beam optic devices are electrostatic to ensure that polarization direction is preserved while the beam is focused. The beam spot at the sample position is a very sensitive to the Einzel lens voltage, magnetic field, and beam energy.

Potential maps, such the one seen in Fig 3.13, are used to predict how the beam will travel through the beamline, and to estimate the tuning parameters of the various focussing elements that will give best beam spot on the sample.

Steering of the beam through the 8 mm square opening in the copper heat shield of the cryostat and focussing of the beam on the sample is done by monitoring the beam spot by using a low-noise CCD camera (Starlight Express, MX516) to image the light emitted when the $^8$Li ions enter a 0.25 mm thick Bicron BC412 plastic scintillator at the sample position. Fig 3.17 of a typical beam spot image shows faintly the 8 mm square window of the cryostat with the beam spot in the center.

The camera is mounted outside the UHV chamber and views the scintillator nearly on the beam axis via a front surface mirror placed upstream that bends the light nearly 90° through a view port. The low dark current of the CCD allows long exposures to be made, with $\sim$10 seconds exposure typically being sufficient to image the beam under normal ion flux conditions. The image may be further analyzed to gain more quantitative information on the beam intensity peak shape. However for tuning the beamline during an experiment, what is of interest is seeing a small, focused beam spot centered on the sample. A surface plot of the entire image can be seen in Fig 3.18.

3.5 $^8$Li stopping distributions and TRIM Calculations

Having knowledge of the $^8$Li$^+$ stopping distribution in the sample is very important to the understanding of the data collected in $\beta$NMR experiments
Figure 3.17: Typical beamspot image of a 5 keV $^8$Li beam focussed on Bicron plastic scintillator taken with Starlight Express CCD camera. 8 mm square hole in the copper heat shield can be seen, as well as the focussed beam spot in the center.[54]

Figure 3.18: Surface plot of beamspot image seen in Fig 3.17.
Figure 3.19: Top: Number of implanted $^8$Li$^+$ as a function of depth from the surface at 500 eV, 5 keV, 10 keV, and 30 keV implantation energies in Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs, generated by Monte-Carlo simulation of 100,000 ions using TRIM.SP software. * denotes 10 keV and 30 keV data scaled by factor of 10 to show detail. Bottom: shows distances further than 1000 Å for highest implantation energy.
Figure 3.20: $^8$Li distribution at various energies, by percent in the layers of Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs, generated by Monte-Carlo simulation of 100,000 ions using TRIM.SP software.
on MML. Since the layers are on the order of just tens of nanometers thick and the hyperfine field at each $^8\text{Li}^+$ depends on the distance away from the magnetic layer that each ion stops from, knowing the relative number of ions stopping in each layer, as well as the stopping distribution profile within the layers is essential in order to analyze the data once its collected. Stopping distributions are generated using the Monte-Carlo simulation software TRIM.SP. This software, developed by W. Eckstein at the Max Planck Institut für Plasmaphysik in Garching,[58] simulates the implantation of a suitable large number of particles, usually on the order of $10^5$. The sample is defined beforehand by entering the thickness, elemental composition, and molar density for each layer. The data that is generated from these simulations is then exported to standard graphing software, such as Origin, where stopping profile as a function of depth can be plotted, and the profile in the layers fit to a Gaussian distribution (see Fig 3.19). TRIM.SP data output also allows one to plot the percentage of $^8\text{Li}^+$ that stops in each layer as a function of implantation energy (Fig 3.20).

In our $\beta$NMR experiments we want to implant at energies such that we are measuring the signal from the Ag layer, so knowing the percentage of $^8\text{Li}^+$ stopping in each layer at a particular energy is required. Knowing the amount of $^8\text{Li}^+$ stopping in different layers for given implantation energy allows us to determine the energies that will result in the maximum number of ions, and therefore maximum signal, in the layer of interest, which is Ag in our case. This information also tells us what signals to expect, as well as the relative intensities. This is extremely important when analyzing the experimental data which may contain $\beta$NMR signals from several different layers of our sample.

A peculiarity that shows up in these calculations is the discontinuity in the stopping distribution at the Ag/Fe interface. The density of Fe ($\rho=7.87 \text{ g/cm}^3$) is lower than that of Ag ($\rho=10.5\text{ g/cm}^3$), so intuitively one would expect a higher stopping power in the Ag than in the Fe, however the spike that is observed in Fig 3.19 suggests otherwise. This disparity may be resolved by examining the electronic structure of Fe compared to Ag. The conduction electron densities in these metals are very different. In units of number($\times 10^{22}$)/cm$^3$ the conduction electron densities are 17.0 for Fe and 5.86 for Ag [59]. At the relatively low implantation energies we use the incident $^8\text{Li}^+$ ions lose energy primarily through ionization of the loosely bound host atoms’ electrons, since the $^8\text{Li}^+$ is not energetic enough to excite core electrons. This effect was confirmed by running TRIM simulations with the
Fe layer replaced by Be and Cs. Be has a very low density ($\rho = 1.85 \text{ g/cm}^3$) but an extremely large valence electron density, ($24.7 \times 10^{22}/\text{cm}^3$), while the Cs has a very similar density ($\rho = 1.87 \text{ g/cm}^3$) but a much smaller valence electron density ($0.91 \times 10^{22}/\text{cm}^3$). The TRIM simulations showed that a large number of $^8\text{Li}$ stopped in the Be layer, while the Cs hardly stopped any at all. This suggests that the observed discontinuity in the stopping distribution at the Ag/Fe interface is a result of the difference in the valence electron density of the two materials.

Once the stopping profiles, $n(x)$, for various implantation energies are generated they can be used with the assumed hyperfine field distribution in Ag, $B(x)$, to generate the distribution of fields, $n(B)$. This is essentially the measurement that is made in a resonance lineshape $\beta$NMR experiment.

### 3.6 Fitting Procedure

With the stopping distributions calculated with TRIM.SP, and the theoretical internal magnetic field distribution, it is possible to generate the model $\beta$NMR lineshape for a particular implantation energy. This lineshape is used in the fitting routine built into Origin 7.5 graphing software and fit to data collected in $\beta$NMR experiments. Origin fits data using a regression method based on the Levenberg-Marquardt (LM) algorithm which is the most widely used algorithm in nonlinear least squares fitting [60]. It is possible to fit several spectra at the same time, and share parameters of the magnetic field distributions. This means we can take several spectra taken at different implantation energies, having different $^8\text{Li}$ stopping distributions, and fit them to the same magnetic distribution. This fitting routine gives the parameters for the induced hyperfine field distribution that brings the model and the experimental spectra into best agreement.

### 3.7 Sample Preparation

Fe/Ag heterostructures studied in these experiments were prepared using molecular beam epitaxial (MBE) at Bret Heinrich’s surface physics laboratory located at Simon Fraser University. A sample is prepared by first loading a GaAs (001) single crystal into an UHV chamber and cleaning the surface by He ion sputtering before thermally annealing the clean surface.
to form large, flat terraces, suitable for preparing Fe/Ag multilayers. The substrate is then moved into the MBE growth chamber where Fe and Ag layers are alternately grown on the substrate by depositing material by thermal evaporation of a heated elemental source sample. Opening a shutter that separates the furnace from the growth chamber exposes the substrate to the evaporated atoms emerging radially from the source and condensing on the substrate surface. The final step is to grow a thin 10 ML Au capping layer that suppresses oxidation of the surface. Film thickness is monitored during growth by counting peaks in reflected high energy electron diffraction (RHEED) oscillations as well as using a quartz crystal monitor, calibrated to take into account of the different subtended solid angle of the sample and quartz monitor. Growth rates were approximately 1-2 ML per minute. The strength of this technique is the constant low energy of the deposited atoms, compared with other techniques, allowing the epitaxial growth of extremely well ordered, crystalline samples with sharp interfaces[61].

In order to produce films with sharp interfaces it is necessary to have close matching of in-plane lattice parameters between the substrate and the epitaxial layer[62]. GaAs(001) is a suitable substrate due to the small mismatch (-1.4%) between the lattice spacing of base-centered cubic (bcc) Fe ($a_{Fe} = 2.87$ Å) and face-centered cubic (fcc) GaAs ($a_{GaAs} = 5.65$ Å), remembering that in GaAs has a 2 atom basis. It is important that the Fe layer is grown first on the GaAs substrate for several reasons. The lattice larger mismatch (2.2%) between GaAs and Ag ($a_{Ag} = 4.09$ Å) leads to a larger strain than in the case of Fe. It has also been found that Ag grows epitaxially in two orientations (001) and (011) on (001)GaAs[64]. GaAs(001) has two different surfaces, either all Ga or As atoms, and for an unreconstructed surface, each surface atom has two unsatisfied bonds oriented along the [110] direction for Ga surface atoms or [110] direction for As surface atoms, see Fig 3.21[63]. The As-stabilized surface leads to (011) Ag orientation, while Ga-stabilized surface gives rise to (001) orientation of Ag[64]. Roughness in real samples means that there will will be domains of Ga and As surfaces, which would lead to domains of different orientation for Ag grown on GaAs, leading to low quality films, while Fe always follows the (001) orientation for both Ga and As terminated surfaces. This, along with the close matching of lattice spacing allows the growth of bbc Fe into very ordered layers[63]. Ag grown on the Fe(001) surface has (001) orientation and fcc structure, with lattice rotated by $\pi/2$[65].

The Ag orientation on the Fe in our samples was confirmed with x-
ray diffraction experiments, which can be seen in Fig 3.22 for the Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs sample. It can be seen that all the major peaks for Ag were parallel to the (001) plane, thus confirming (001) orientation of Ag. The “satellite” peaks that can be seen on either side of the Ag(002) peak can be accounted for by considering the finite thickness effect of the Ag layer[66]. Similar peaks for the smaller Ag(004) peak cannot be distinguished from the noise. Since there are only 14 monolayers (ML) of Fe, compared with 400 ML of Ag in the sample, and just 20 ML of Au on the surface, the signal peaks from the Fe and Au are very difficult to pick out from the noise, so it isn’t possible to make any conclusive arguments about the structure of these layers from the x-ray diffraction, however their (001) orientation was confirmed by RHEED measurements during growth.
Figure 3.21: Schematic diagram showing how the unsatisfied surface bond orbitals are distinguished in unreconstructed GaAs(001) for Ga(001) and As(001) terminated surfaces. Roughness in real samples will have domains of Ga and As, making growth of well ordered Ag films directly on GaAs difficult[63].
Figure 3.22: XRD spectrum of Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs showing peaks from planes parallel to (001) in both Ag and GaAs, confirming their (001) orientation.
Chapter 4

Results

The stopping distribution of $^8$Li is a crucial part of the procedure to extract the spatial dependence of the hyperfine fields in the Ag spacer layer in a magnetic multilayer. It is therefore necessary to verify that the results of the TRIM.SP simulations for implantation match the $\beta$NMR data collected at various implantation energies. In this section we will review the results obtained from TRIM.SP calculations and how they are used in the analysis. Then results of fitting the model lineshape to $\beta$NMR spectra will be presented, along with the parameters for the spatial dependence of the induced hyperfine fields in the Ag layer.

4.1 TRIM.SP results

In order to verify that the implantation simulations produced using the TRIM.SP software are accurate we compare the predicted stopping distributions for various energies of ion implantation with the $\beta$NMR spectra taken in two different MML samples, Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs, and Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs. Based on knowledge gained in previous experiments to characterize the $\beta$NMR resonances of $^8$Li in the various layers, Au [55], Ag [56], and GaAs [67] we determine the relative number of $^8$Li that stop in each layer from the relative amplitude of each layers’ characteristic peak, and compare this to the distribution that results from the TRIM simulation at the same energy.

At room temperature the implanted $^8$Li stops primarily at a substitional site in both Au and Ag resulting in very narrow $\beta$NMR resonances that have intrinsic linewidths on the order of just a few hundred Hz. These resonant lines are also shifted by a small characteristic amount as a result of the electronic environment within the sample. This is called the Knight shift and is a result of the small polarization in conduction electrons at the Fermi surface of metals due to the presence of the external magnetic field. The hyperfine interaction between the polarized electrons and the $^8$Li gives rise
to The Knight shifts in Au and Ag. Measured relative to insulator MgO resonance, these shifts are +60(20) and +120(12) ppm respectively[55]. The $\beta$NMR resonance that is measured in the semiconductor substrate GaAs, has no Knight shift, but is considerably broader (3-4 kHz) than either Au or Ag due to the natural abundance of sizable nuclear moments of $^{69}$Ga, $^{71}$Ga, and $^{75}$As isotopes [67]. The large internal hyperfine fields experienced by the $^8$Li stopping in the Fe layer implies that the resonance in Fe is well outside the frequency range used in these experiments. Furthermore, any $^8$Li that stops in Fe at room temperature shows a very fast spin-lattice relaxation rate relative to the $^8$Li half life due to scattering of magnetic excitations in the Fe. This will have the effect of reducing the off resonance asymmetry.

Fig 4.1 shows the $\beta$NMR spectra taken in the Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs sample at four energies of implantation between 500 eV and 30 keV. The spectra, which have been normalized to the off resonance amplitude, $A_0$, were carried out at room temperature ($\sim$285 K) in a longitudinal field of $H_0 \simeq 4.1$ T. The stopping profiles, calculated with TRIM.SP at these same four energies can be seen in Fig 4.2. Fig 4.3 shows the percentage of implanted $^8$Li that stops in a particular layer as a function of energy for this sample.

The main resonant peak of the first spectrum, Fig 4.1(a), is most prominent in only the spectrum taken at the lowest energy of implantation, $E=500$ eV. This peak is shifted by $\sim$+80 ppm relative to the GaAs, which is in good agreement with the Knight shift measured previously for Au, and is therefore attributed to most of the $^8$Li stopping in the 40 Å Au capping layer. This is supported by the TRIM.SP results that show nearly all of the $^8$Li stopping in this layer at this energy, Figs 4.2(a) and 4.3. This resonant peak, attributed to Au, diminishes rapidly for implantation energies above 500 eV, also predicted by the TRIM.SP results. Also a second line, with larger frequency shift than the Au, increases at these energies, Fig 4.1(b). This peak is shifted by $\sim$120 ppm relative to the GaAs, in very good agreement with the Knight shift measured in Ag. This narrow line has a width that is comparable to the Ag line observed in Ag films grown on nonmagnetic substrates, which would indicate that most of the $^8$Li is stopping in the Ag close to the Au/Ag interface farthest from the Fe layer where the hyperfine coupling to the magnetic Fe is negligibly small. However this line does appear to have a broad base resulting from the hyperfine fields experienced by the small amount of $^8$Li that stops close to the Ag/Fe interface. This situation is supported by the TRIM.SP stopping profile for implantation energy of 5 keV, Fig 4.2(b), which shows that $^8$Li is predominantly located in the Ag layer close to the
Figure 4.1: $\beta$ spectra taken at room temperature in a longitudinal field of $H_0 \sim 4.1$ T in Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs at (a) 500 eV, (b) 5 keV, (c) 10 keV, and (d) 30 keV implantation energy. The error bars in the spectra are statistical, but the noise in the spectra is systematic.
Figure 4.2: TRIM.SP calculated stopping distributions in Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs at energies (a) 500 eV, (b) 5 keV, (c) 10 keV, and (d) 30 keV.
Figure 4.3: TRIM.SP calculated stopping distributions in each layer as a function of energy for Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs.
Au (400 Å from the Fe layer).

As the implantation energy is increased, the $^8$Li ions stop deeper into the sample and therefore closer to the Fe/Ag interface. At 10 keV, Fig 4.1(c) shows the Au line now completely gone, and the Ag line has been significantly broadened at frequencies on both the high and low frequency sides, suggesting the presence of large positive and negative hyperfine fields in this region. The peak also shows asymmetry which could be the result of an emerging GaAs peak at lower frequencies. Again, TRIM.SP profiles calculated at E=10 keV implantation, Fig 4.2(c), supports this interpretation. It can be seen that most of the $^8$Li stops in the region within 400 Å of the Fe/Ag interface, as well as a small amount ending up in the GaAs substrate.

At full implantation energy, E=30 keV, a broad peak is observed that we attribute to the GaAs substrate, Fig 4.1(d) since the TRIM.SP calculations for full implantation energy shows that the majority of the $^8$Li stops in the GaAs substrate, Fig 4.2, with some still stopping in the Ag close to the Fe, which would account for the asymmetry in this line, with more weight on the high frequency side of the spectrum.

A second sample (Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs) was also compared to TRIM.SP calculated distributions. Experiments on this sample were also conducted at room temperature (∼285 K), but the longitudinal field was $H_0=4.5$ T, which is why the resonances are at a higher frequencies in Fig 4.4 compared to Fig 4.1. These spectra have not been normalized by $A_0$ for reasons that will be explained later. The TRIM.SP calculated distributions in this sample are shown in Figs 4.5 and 4.6.

The $\beta$NMR spectra taken at 2.5 and 5.5 keV (see Fig 4.4) both show a broadened Ag line, with the 2.5 keV line being slight asymmetric due a small Au line at lower frequency. The TRIM.SP calculated profiles, see Fig 4.5 of this this energy range show that the $^8$Li is primarily stopping in the 200 Å Ag layer at these two energies, closer to the Au/Ag interface at 2.5 keV, and the Ag/Fe interface at 5.5 keV as expected. It can be seen in Fig 4.6 that in this sample, with a much thicker Fe layer than the previous sample, that quite a bit of $^8$Li stops in the Fe layer at 5.5 keV, compared with 2.5 keV. However this has little effect on the shape of the spectrum. While the spectra in Fig 4.1 were normalized by the off resonance asymmetry, the spectra in Fig 4.4 were not normalized to show the loss in off resonance asymmetry, $A_0$. One can see how $A_0$ is reduced at 5.5 keV from the fast relaxation of $^8$Li stopping in and near the Fe layer. At the full implantation energy of 29.5
Figure 4.4: \( \beta \)NMR spectrum taken at room temperature in magnetic field \( H_0 \sim 4.5 \text{T} \) in \( \text{Au}(40 \text{ Å})/\text{Ag}(200 \text{ Å})/\text{Fe}(140 \text{ Å})/\text{GaAs} \) at implantation energies (a) 2.5 keV, (b) 5.5 keV, (c) 9.5 keV, and (d) 29.5 keV. The error bars in the spectra are statistical, but the noise in the spectra is systematic.
Figure 4.5: TRIM.SP calculated stopping distributions in Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs for implantation energies (a) 2.5 keV, (b) 5.5 keV, (c) 9.5 keV, and (d) 29.5 keV. Inset: shows further range into substrate for highest implantation energies.
Figure 4.6: TRIM.SP calculations of percentance of implanted $^8$Li stopping in each layer as function of energy in Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs.
keV we see a very narrow GaAs that comes from $^8\text{Li}$ stopping deep within the substrate, inset Fig 4.4. At implantation energies around 9.5 keV, the βNMR spectrum has an intermediate appearance between the spectra of 5.5 and 29.5 keV. It can be seen, Fig 4.6, that implantation energies close to 10 keV results in nearly equal distribution between the Ag, Fe, and GaAs layers. Since we get no signal from Fe, we would expect that the signal would be almost equal parts GaAs substrate and Ag.

As was shown in Chapter 2, in many cases the resonance lines can be described by a Lorentzian shape:

$$A(\omega) = A_0 - A_{RF}(0) \frac{\Delta}{(\omega_0 - \omega)^2 + \Delta^2}$$

where $A_0$ is the off resonance asymmetry and $\Delta$ is the half width at half the maximum amplitude, and $\omega_0$ is the resonance frequency.

Although the Lorentzian fits do not provide details about the decay of induced hyperfine magnetism away from the magnetic/nonmagnetic interface, it does provide an easy way to follow general trends in the data. The width of the Lorentzian gives us an idea of the average induced hyperfine fields in the Ag layer. It makes sense that larger distributions of induced magnetism will give rise to broader distributions. The area of the Lorentzian gives us a measure of the total signal strength, so if we really are losing signal from fast relaxation of $^8\text{Li}$ stopping in Fe we should see a minimum in the signal strength at the maximum implantation energy.

Fig 4.7 shows the area under the Lorentzian fits to the resonance line which shows a minimum in the signal for implantation energies around 5 keV. TRIM.SP calculations predicts the maximum amount of $^8\text{Li}$ stopping at the Fe layer at slightly higher implantation energy of $\sim 7.5$ keV, Fig 4.6. This could be interpreted as the TRIM.SP giving slightly shallower implantation at a particular energy than is observed. It has been observed in previous studies that the TRIM.SP calculations predict slightly deeper implantation for $^8\text{Li}$ at similar energies than is observed[68]. It is also likely that $^8\text{Li}$ stopping in the Ag very close to the Ag/Fe interface also relaxes faster than in Ag away from the interface. This would also lead to a slightly lower value for the implantation energy of the minimum in signal strength.

These results qualitatively confirm that the Monte-Carlo TRIM.SP software accurately calculates the $^8\text{Li}$ stopping distribution in this sample. It is therefore reasonable to use the results of these calculations as the stopping profile of $^8\text{Li}$ in the Ag layer in the modeling of our βNMR lineshape.
Figure 4.7: The area under the Lorentzian fits of $\beta$NMR resonances lines, showing a minimum at implantation energy of $\sim$5 keV, resulting from fast $^8$Li relaxation in the presence of large hyperfine fields in and close to the magnetic Fe layer.
as discussed in Chapter 2. It also indicates the presence of large positive and negative hyperfine fields induced in the Ag close to the Fe, but from the spectra taken in the 800 Å thick Ag sample, these induced fields become negligible at distances greater than approximately 400 Å away from the Fe. In this region, far from the magnetic layer, the spectra appear to have very similar widths to spectra taken in Ag thin films grown on a non-magnetic substrate at room temperature, Fig 3.16.

In order to use the fitting procedure described in chapter 3 to extract the parameters of the hyperfine coupling in the Ag layer, we need spectra that have been taken with implantation energies such that all, or most of the signal comes from $^8\text{Li}$ stopping in the Ag close to the Fe layer. Unfortunately at higher implantation energies the $^8\text{Li}$ range straggling is largest. This presents a problem in the Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs sample, where at energies sufficiently high to stop an appreciable amount of $^8\text{Li}$ in the region close to the Ag/Fe interface, a significant amount of $^8\text{Li}$ stops in the GaAs substrate. This distorts the signal and makes fitting to our model difficult. The Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs sample is more promising. Since the Fe layer is much thicker and the Ag is thinner most of the observed signal comes from the Ag close to the interface. Implantation energies of 2.5 and 3.0 keV were determined to be the best spectra, since signal from the Au layer become significant at implantation energies lower than 2.5 keV, and the GaAs substrate has already become significant at implantation energy of 5 keV. The stopping distributions calculated for these energies with TRIM.SP were fit in just the Ag layer to a Gaussian distribution, to determine the center and width of the stopping profile. The results of this fit can be seen in Fig 4.8, and the values obtained are given in Table 4.1.

Using these values for the stopping distribution, the two spectra were fit together to the function we derived in Chapter 2:

$$A = A_0(1 - an(\omega))$$

where $n(\omega)$ is based on the Gaussian stopping distribution and the distribution of magnetic fields:

$$B_{\text{max}}(x) = B_{\text{ext}} + \frac{B_0}{1 + (x/\lambda_F)^\alpha};$$

$$B_{\text{min}}(x) = B_{\text{ext}} - \frac{B_0}{1 + (x/\lambda_F)^\alpha}$$
with the free parameters \( \nu_0 \) (which is related to \( B_0 \) by \( \gamma \)) and \( \alpha \) being shared, while \( \nu_{\text{ext}} \) (which is related by \( \gamma \) to \( B_{\text{ext}} \)), \( a \), and \( A_0 \) were independent for each energy. The result of the best fit can be seen in Fig 4.9.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Center(Å)</th>
<th>Width(Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.5</td>
<td>160</td>
<td>168</td>
</tr>
<tr>
<td>3.0</td>
<td>145</td>
<td>190</td>
</tr>
</tbody>
</table>

Table 4.1: Gaussian fit parameters of TRIM.SP stopping profiles at 2.5 and 3.0 keV implantation energy in the Ag layer of Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs sample.

<table>
<thead>
<tr>
<th>Energy</th>
<th>( \alpha ) (shared)</th>
<th>( \nu_0 ) (kHz) (shared)</th>
<th>( a \times 10^{-5} )</th>
<th>( A_0 )</th>
<th>( \nu_{\text{ext}} ) (kHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.5 keV</td>
<td>1.84 ±0.05</td>
<td>1942 ±266</td>
<td>(3 ±0.4)</td>
<td>0.1236 ±0.0001</td>
<td>28358 ±18</td>
</tr>
<tr>
<td>3.0 keV</td>
<td>1.84 ±0.05</td>
<td>1942 ±266</td>
<td>(2 ±0.3)</td>
<td>0.1277 ±0.0001</td>
<td>28363 ±22</td>
</tr>
</tbody>
</table>

Table 4.2: Free parameters values obtained from fitting the theoretical lineshape to resonant lines measured in Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs at implantation energies 2.5 and 3.0 keV, room temperature, and \( H_0 \sim 4.5 \) T.

Dividing \( \nu_0 \) by \( \gamma=6301 \) kHz/T gives the result \( B_0 = 0.38 \pm 0.04 \) T for the hyperfine coupling of \(^8\text{Li}\) at the Fe/Ag interface. This is fortuitously close the calculated value of 0.300 T [40] for the induced hyperfine field at the first Ag layer from the Fe/Ag interface. The decay constant, \( \alpha=1.84 \pm 0.05 \), agrees reasonably well with the theoretical value of 2, however is much larger than the values \( \alpha=0.4(1) \) and 0.8(1) measured in Fe(40 Å)/Ag(3000 Å)(001) and Fe(40 Å)/Ag(200 Å)/Fe(40 Å)(001) samples, respectively, using the complementary technique of Low Energy Muon Spin Resonance (LE-\( \mu \)SR) [19, 69].

The spectra in Fig 4.9 show a sharp peak that is missed by the fitting function, which shows a “flat top”. The flat top of the fitting function indicates that the induced hyperfine fields have not completely decayed away in the Ag furthest from the Fe, which is 200 Å in this case. The sharp peak that is missed is most likely due to the small amount of Li that stops in the Au cap at these energies, as seen in the Figs 4.2 & 4.3.
Figure 4.8: Gaussian fit (dotted line) to TRIM.SP calculated distributions (solid line) in the Ag layer of Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs for 2.5 keV and 3.0 keV implantation energies.
Figure 4.9: Fit of the theoretical lineshape (solid line) to βNMR spectra (points) taken at room temperature and $H_0 \sim 4.5$ T in Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs for 2.5 keV and 3.0 keV implantation energies.
Figure 4.10: Form of the induced hyperfine fields in the Ag layer as determined from the fitting procedure. *Inset* shows expanded distance to show detain far from the Fe/Ag interface.
This form for the induced hyperfine fields can be seen in Fig 4.10. It shows that the induced fields are fairly large near the Fe/Ag interface, but the inset shows that the induced magnetism is very small at distances of $\sim 400 \, \text{Å}$, as was observed in the Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs sample.
Chapter 5

Conclusion

The results of the experiments on two Fe/Ag heterostructures, Au(40 Å)/Ag(800 Å)/Fe(20 Å)/GaAs and Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs, show that $\beta$NMR is well suited to studying the induced internal magnetism in MML structures. We have shown that by controlling the implantation energy of the radioactive $^8$Li$^+$ NMR probes, it is possible to profile the internal magnetic environment of thin film samples from within 40 Å from the sample surface to depths of 2000 Å, which is well into the sample bulk. We have demonstrated that $\beta$NMR is capable of measuring the internal magnetic environment of these magnetic/nonmagnetic thin films, and will be a very useful tool as industry begins to manufacture devices on nanoscopic scales, and particularly with the spin properties of charge carriers becoming utilized in spintronic devices.

We have introduced a model $\beta$NMR lineshape based on the theories of induced hyperfine coupling into a nonmagnetic layer adjacent to magnetic layer. These theories were developed by extending the RKKY theory describing the spin polarization in the host metal’s conduction electrons in the region of a magnetic impurity atom. We anticipated that the coherent oscillations of the electronic polarization in the nonmagnetic layer should be wiped out over the lateral distances of a few nm determined by the beam spot of the $^8$Li beam due to roughness at the interface. The result of this is that we don’t measure the sharp features characteristic of coherent oscillations, and that the hyperfine magnetism is instead “smeared out”. The model lineshape was found to fit very well to $\beta$NMR resonances measured by implanting $^8$Li into the Ag layer of of a Au(40 Å)/Ag(200 Å)/Fe(140 Å)/GaAs sample at energies 2.5 and 3.0 keV, which confirmed our prediction for the form of the induced hyperfine fields, and yielded values for the hyperfine distribution. The fits show that the value of the maximum hyperfine fields at the Ag/Fe interface is $B_0=0.38 \pm 0.04$ T, and that the induced magnetism decays away from the interface following the power law $(x/\lambda_F)^{1.84\pm0.05}$. The hyperfine distribution that was extracted from the fits was shown in Fig 4.10. The
hyperfine field at the interface agrees well with theoretical predictions [40], as well as experimental values for the hyperfine coupling of Li in Fe [41]. Some ways to improve the method of measuring the induced magnetism were discovered during the course of these experiments. It can be seen from the TRIM.SP calculated stopping distributions that as the energy of implantation is increased, the width of the distribution also increases. In the samples examined in these experiments, the Ag/Fe interface, which is the region of interest, was 240 Å and 840 Å from the sample surface. The implantation energy required to implant $^8$Li in this region results in a stopping distribution that is very wide. This wide distribution makes it difficult to obtain a clean signal from just Ag close to the Fe/Ag interface since much of the $^8$Li is being stopped in other layers such as the GaAs substrate. To get around this problem we have several samples of varying Ag thicknesses of 50Å, 100Å, and 150Å, but identical Fe and Au layers between samples. This would allow us to implant at low energies (∼1 keV) in all samples, which would result in a very narrow stopping distribution but still allow depth profiling of the induced hyperfine magnetism. This will have the advantage of sampling very narrow slices of the induced magnetism in only the Ag layer at various distances away from the Fe/Ag interface, which we expect will allow better characterization of the spatial dependence of the induced magnetism.

Our measurements could also be improved by using a new experimental mode that allows us to measure resonances using pulsed RF that was not available at the time the experiments presented in this work were carried out. This pulsed RF mode gives a better measurement of the lineshape by avoiding the dynamic and systematic effects that are present with continuous RF, which would lead to a better measurement of the induced hyperfine magnetism in these samples.
Bibliography


[54] Figure taken from http://bnmr.triumf.ca/


[57] http://bnmr.triumf.ca/


