Experiment 815, 816, 817, 897 and 913: β-NMR

(W.A. MacFarlane, UBC Chemistry for the β-NMR group)

The development of β-NMR at ISAC is one of the major efforts at TRIUMF to employ radioactive ion beams in the fields of condensed matter and materials science. These experiments use implanted beta radioactive ions as NMR (nuclear magnetic resonance) probes of local magnetic fields in materials. The beam energies at ISAC are such that 1) the ions penetrate at most a few hundred nanometres (nm) into conventional solids and 2) it is possible to electrostatically decelerate the incident beam (by biasing the target at a high voltage) to vary the implantation depth down to the level of a few nm. Thus many technologically relevant and scientifically interesting phenomena occuring near surfaces and in synthetic thin film heterostructures can be studied with β-NMR. Moreover, there are very few competing depth-resolved probes of such effects. The β-NMR group is pursuing these questions in a variety of materials under several TRIUMF EEC proposals. I highlight some of the recent results in these areas below. It should be noted that simultaneously with these measurements, we have been making important technical advances in the instrumentation in order to carry out these measurements with the limited beamtime available at ISAC, some of these are detailed in the last section.

0.0.1 Conventional Metals

As part of the initial testing and commissioning of the β-NMR spectrometer at ISAC, we have studied some simple elemental metals, for example silver (see publication 2). These measurements are important in establishing the behaviour of the probe in simple materials, for example. There is also fundamental interest in these materials (see **E815** and the new TRIUMF EEC proposal **E1042**). As part of this ongoing effort we recently studied a thin film of copper. The behaviour is very similar to that of Au and Ag (reported previously), but the resonances are broader, due to the copper nuclear magnetic dipoles. Similar to Au and Ag, we find a transition from a cubic interstitial site for the implanted ^{8}Li at low temperature to a substitutional site at higher temperature. The transition occurs at a slightly lower temperature than for Ag - see Fig. 1.

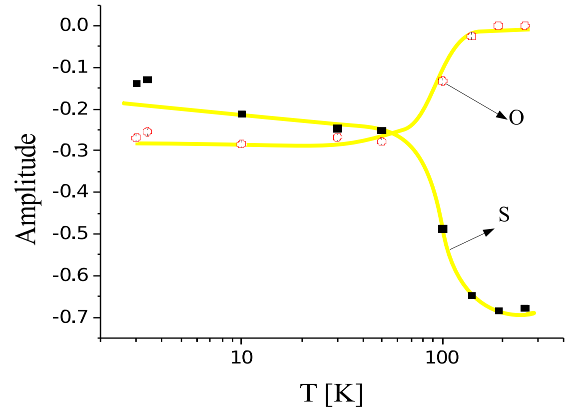

Fig. 1: The resonance amplitudes of the ^{8}Li implanted in a thin Cu film as a function of temperature reflecting the site transition above 100 K, from the octahedral (O) interstitial site to the substitutional (S) site.

Palladium is another cubic close packed elemental metal which we have previously used to measure ^{8}Li spin relaxation. We have now extended these measurements to the resonances, where we find a remarkably large and negative frequency shift, see Fig. 2. This behaviour is distinct from all the other simple metals we have measured so far and is the subject of active investigation (**E1042**).

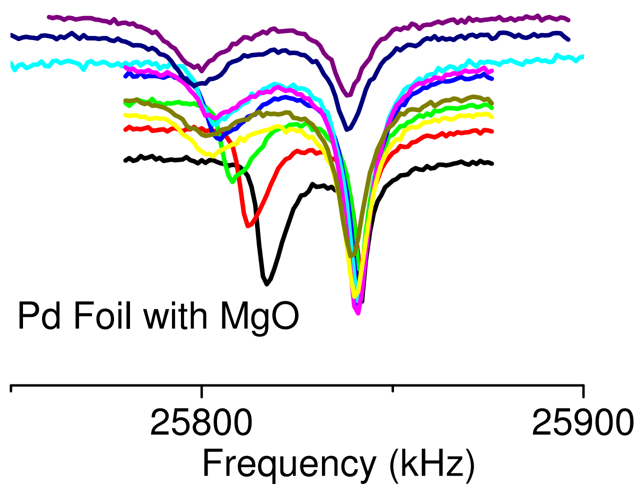

Fig. 2: The resonances of ^{8}Li implanted in a thin foil of Pd as a function of temperature. The foil was wrapped around a crystalline substrate of MgO and pierced with a pin. Centering the beamspot on the pinhole resulted in the composite spectrum including the MgO resonance at the right of the broader Pd resonance.

Niobium is an elemental superconducting metal with the body-centred cubic structure which differs from the cubic close packed structure in that there are no interstital sites of cubic symmetry. Like the other elemental metals we find that the ^{8}Li stops in an interstitial site at low temperature, and at higher temperature it moves to a substitutional site. In Niobium this situation is dramatically confirmed as the sites differ in symmetry, and, due to the electric quadrupole moment of the ^{8}Li nucleus, the resonance of the interstitial site is split into quadrupolar satellites - see Fig. 3. This demonstrates the potential to use the quadrupole interaction to identify the ^{8}Li site.

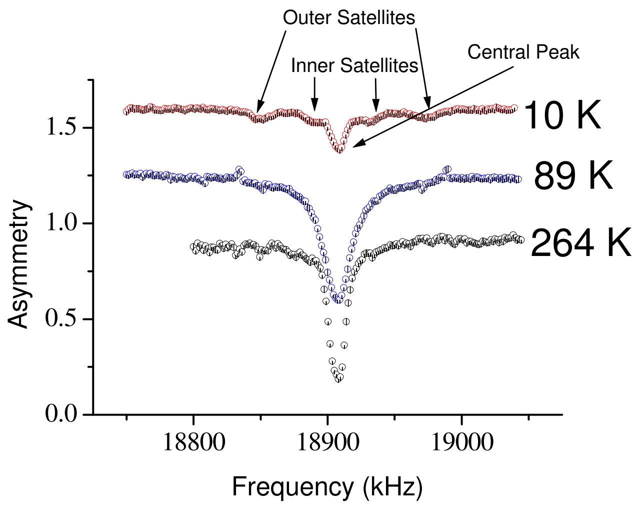

Fig. 3: The resonance of ^{8}Li implanted in an oriented thin film of Niobium on a sapphire substrate.

0.0.2 Magnetic Heterostructures

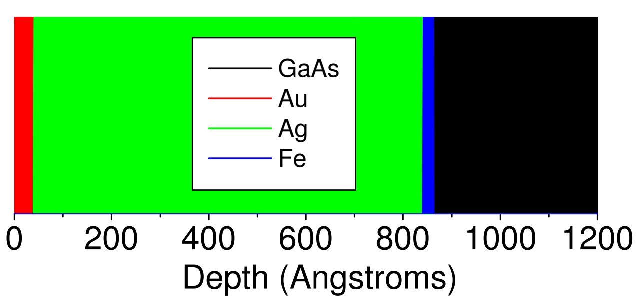

Fig. 4: A schematic of the heterostructure for which a platform bias scan is shown in the following figure. The thicknesses are to scale.

As part of **E815**, we have been studying a variety of epitaxial metallic heterostructures of iron and silver on GaAs substrates. These kinds of structures are important as they can exhibit giant magnetoresistance and are for this reason used in magnetic storage applications. The effect is a consequence of the depth-dependent magnetic polarization of the nonmagnetic layer by the ferromagnetic layer. A detailed understanding of this polarization (which would represent a significant advance in the field) - requires a depth sensitive magnetic probe such as β-NMR.

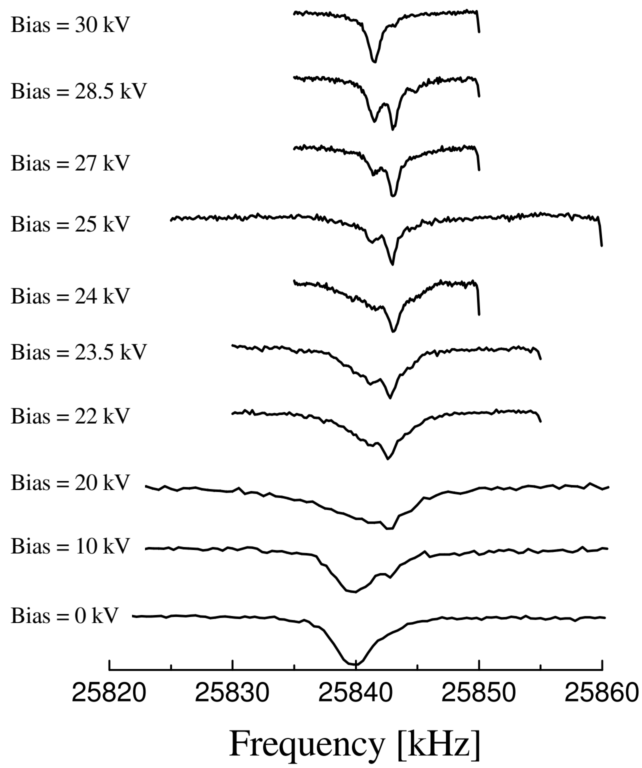

Fig. 5: A bias scan of the ^{8}Li resonances in the heterostructure of Fig. 4 at 280 K. As the bias goes up the implantation energy goes down. The change in the resonance with bias thus demonstrates the depth resolution of the technique.

We have made preliminary studies on several structures of this type, one of which is shown schematically in Fig. 4. This structure had a thick Ag layer deposited on top of a thin magnetic Fe layer. The Ag is then capped with 20 monolayers (4 nm) of Au to protect it. In order to vary the implantation depth the sample was biased to a series of positive voltages. The implantation energy is thus the beam energy minus this bias voltage. In Fig. 5, the resonances are shown at a temperature of 280 K as a function of the bias voltage. The changes in the resonance as a function of voltage clearly show the depth sensitvity of this technique. In particular, at 25 kV the signal is primarily that of unperturbed Ag. At this bias the stopping energy (about 600 eV) is such that Li stops mainly in the Ag which is too far from the Fe to be perturbed. At the maximum bias (about 100 eV implantation energy), the signal is nearly that of pure gold, i.e. we are stopping all the probe ions in a 4nm overlayer! This sample is not the ideal geometry to study the polarizing effects of the Fe film however, so other samples have been and will be studied. One example is shown in Fig. 6. The structure is analogous to that shown in Fig. 4, except the Ag layer is only 20 nm thick. Measurements were taken at room temperature, in an applied external field of 4.5 T, for a range of implantation energies where most of the ^{8}Li stops within the Ag layer. The distribution of magnetic fields in the Ag layer is reflected in the resonance lineshapes in Fig. 6. The hyperfine coupling is predicted to have an oscillating spatial dependence for a perfectly sharp interface, however roughness at the interface will introduce random phases that will tend to suppress these oscillations upon averaging over a large lateral area determined by the beam spot, several mm in diameter. Using the stopping distribution of ^{8}Li in the sample, and the form of the hyperfine field decay, it was possible to model the resonance and extract the form of the hyperfine field decay in the Ag layer, which was determined to be a power law $x^{-1.2}$. Detailed analysis is ongoing and will be used to compare with theory and other types of measurements.

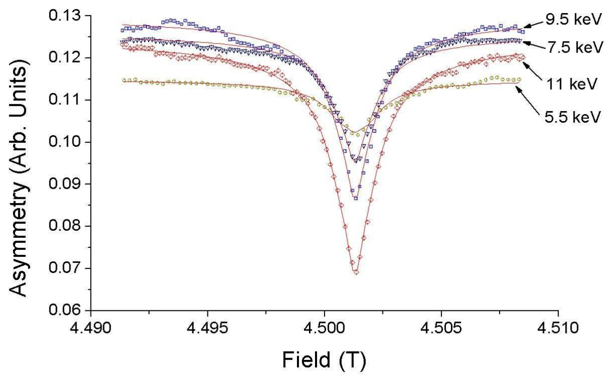

Fig. 6: The resonances as a function of implantation in energy in a thin Fe/Ag heterostructure (see text) - courtesy of T.A. Keeler

0.0.3 Semiconductors

As part of **E816** and **E913** we have been studying high purity crystalline semiconductors. Here the implanted lithium is interesting as an electronically active impurity. Also since these materials are often used as substrates (e.g. for the magnetic heterostructures above) a proper characterization of the β-NMR will be very important for interpreting such measurements correctly.

We have made initial studies of the structure and dynamics associated with ^{8}Li$^+$ in the semiconductors GaAs, Si and Ge. In GaAs (as previously reported) at low temperatures, the amplitude of the ^{8}Li$^+$ resonance indicates that a large fraction (at least 70%) of the Li end up in locations with cubic symmetry (such as the tetrahedral interstitial and substitutional sites). The linewidth of the β-NMR ^{8}Li$^+$ resonance increases dramatically above 150 K, reaches a maximum at about 290 K, and decreases again. This suggests a site-change transition for Li, probably from an interstitial to a substitutional site, at $\approx$ 150 K, similar to what is found in metals. The picture is similar in Germanium, but the line is much narrower at all temperatures, exhibiting only a slight broadening up to room temperature. Additionally, at room T there appears a small satellite line shifted to high frequency from the main resonance - see Fig. 7.

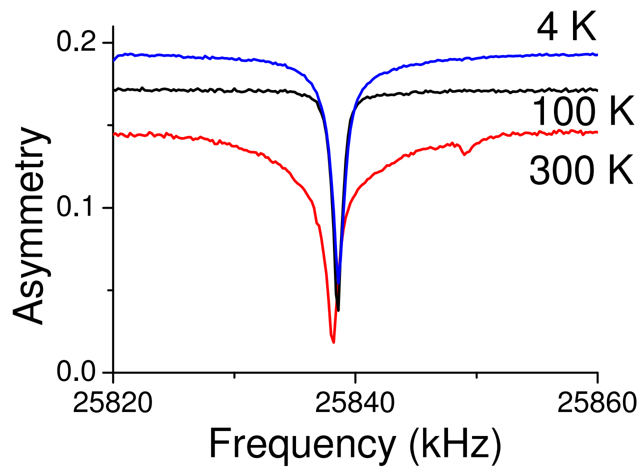

Fig. 7: The resonance of ^{8}Li implanted in a crystal of Ge as a function of temperature.

0.0.4 Superconductors

The magnetic field distribution in $NbSe_2$ was measured using β-detected NMR of ^{8}Li with a variable implantation energy of 1-30 keV. Figure 8 below shows the β-NMR lineshape above and below the superconducting transition temperature (T_c=7.0K) in a magnetic field of 0.3T parallel to the hexagonal c-axis. In the normal state at T=10K a symmetric lineshape is observed with a linewidth attributed to nuclear dipolar broadening from the Nb nuclear moments. Surprisingly there is no evidence for a quadrupolar splitting in the resonance even though $NbSe_2$ is noncubic. At T=0.5Tc (3.5K) the characteristic lineshape from a vortex lattice is clearly evident; the peak frequency, from the region between vortices, shifts to a slightly lower frequency and a high frequency develops from the vortex core region. The lineshape was fitted to a modified London model with a Gaussian cutoff function yielding a penetration depth of 272 nm and a coherence length of 12 nm which can be compared with μSR measurements from deeper inside the material. The dependence on implantation energy has also been studied.

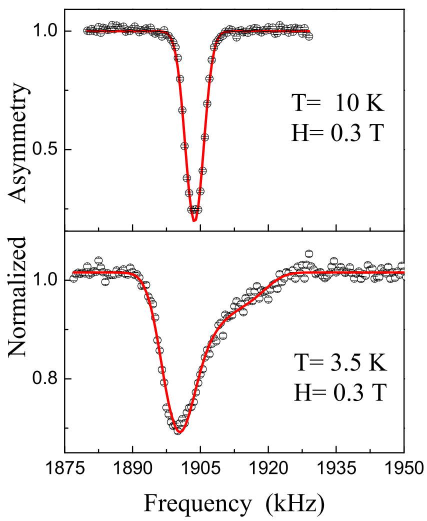

Fig. 8: Beta-detected nuclear magnetic resonances in $NbSe_2$ above and below the superconducting transition temperature of 7.0K

While $NbSe_2$ is a very well studied conventional superconductor, it's transition temperature is quite low. The recent discovery of high temperature "conventional" superconductivity in MgB_2 may provide another important test case. A preliminary room temperature resonance in a thin film of hexagonal MgB_2 on a sapphire substrate is shown in Fig. 9. The resonance is broad but shows no large quadrupolar effects.

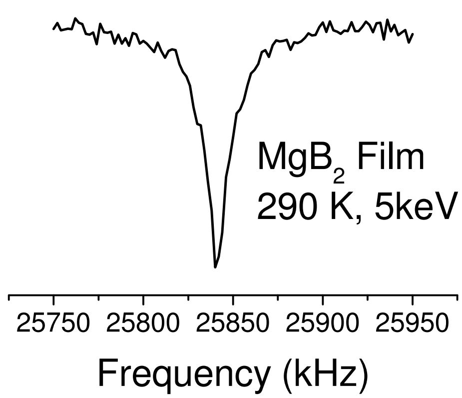

Fig. 9: The resonance lineshape of ^{8}Li implanted in a think film of MgB_2 deopsited on a sapphire substrate.

0.0.5 Beta Detected Nuclear Quadrupole Resonance

As previously reported, the first beta-detected nuclear quadrupole resonances (β-NQR) of ^{8}Li at zero field were observed using the newly developed βNQR spectrometer (see publication 3). The resonances were detected in $SrTiO_3$, Al_2O_3 and Sr_2RuO_4 single crystals by monitoring the beta-decay anisotropy as a function of a small audio frequency magnetic field (see figure 10 below). The resonances show clearly that ^{8}Li occupies one site with non-cubic symmetry in $SrTiO_3$, two in Al_2O_3 and three sites in Sr_2RuO_4.

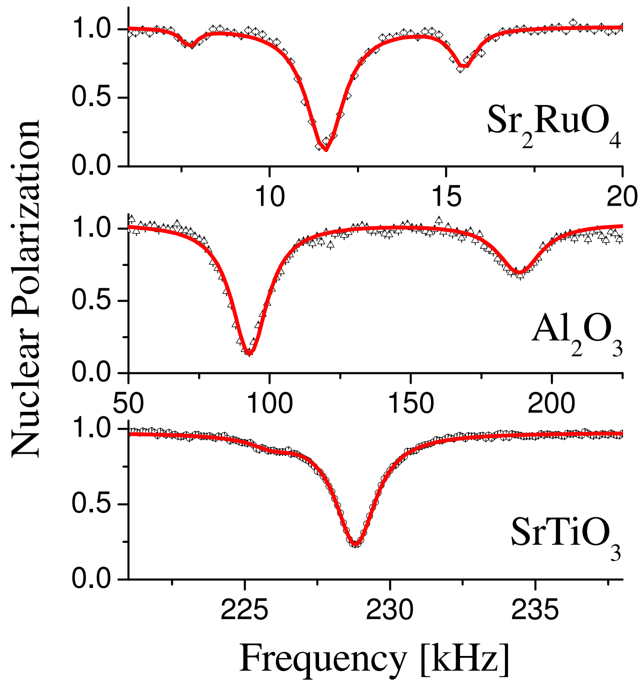

Fig. 10: Beta-detected nuclear quadrupole resonances in $SrTiO_3$, Al_2O_3 and Sr_2RuO_4 single crystals

The resonance amplitude and width are surprisingly large compared to the values expected due to the transition between $2 \rightarrow 1$ spin states of the ^{8}Li nuclei. The main reasons for the enhanced amplitudes is the effect of small non-axial electric field gradient, which mixes the ± 2 and the ± 1 states, enabling a larger loss of the polarization compared to that expected with vanishing non-axial electric field gradient (i.e. without mixing of the different spin states).

Zero field μSR measurements have proven to be very useful in studies of magnetism. Similarly, we expect that zero field β-NQR will provide a very useful addition for studies of magnetism in thin films and nano-structures.

0.0.6 Summary of Technical Advances

We have in the past several years transformed the polarimeter station into the βNQR spectrometer, a second instrument capable of measurements in low fields up to about 150 Gauss. This spectrometer is equipped with its own cryostat, pulsed radiofrequency(RF) magnetic system, and we are in the process of designing and implementing a deceleration system similar to the high field platform. We are moving towards near-simultaneous operation of the two spectrometers. For polarization monitoring, we have implemented the "Neutral Beam Monitor" which intercepts the neutral ^{8}Li atoms that are not reionized in the Helium vapour cell at the end of the polarizer. Numerous advances have also been made in the data acquisition software, laser polarizer, detector and RF components of the spectrometer. We have also identified problems with ISAC beam stability as well as advanced our understanding of the beam dynamics near the sample. We have in-situ capability to monitor the beamspot, using a sapphire scintillating crystal and a CCD camera. We identified a problem with discharge electrons produced in the final Einzel lens in the beamline being accelerated and focused by the high magnetic field in the β-NMR spectrometer. We have recently addressed this problem with a negative ring barrier electrode in consultation with M. Olivo. Technical advances such as these will allow us to make maximal use of the scarce ISAC beamtime.

0.0.7 Publications

1. "Application of Low Energy Spin Polarized Radioactive Ion Beams in Condensed Matter Research", R.F. Kiefl *et al.*, Nuclear Physics News, Vol. 15 No. 1, 2005.

2. "β detected NMR of ^{8}Li implanted in a thin silver film", G.D. Morris, W.A. MacFarlane, K.H. Chow, Z. Salman, D.J. Arseneau, S. Daviel, A. Hatakeyama, S.R. Kreitzman, C.D.P. Levy, R. Poutissou, R.H. Heffner, J.E.Elenewski, L.H. Greene and R.F. Kiefl, Physical Review Letters, **93** 157601 (2004).

3. "Beta-Detected Nuclear Quadrupole Resonance with a Low Energy Beam of ^{8}Li$^+$", Z. Salman, E.P. Reynard, W.A. MacFarlane, J. Chakhalian, K.H. Chow, S. Kreitzman, S. Daviel, C.D.P. Levy R. Poutissou and R.F. Kiefl, Physical Review B **70** 104404 (2004).

0.0.8 Invited Talks

1. R.F. Kiefl: Canadian Association of Physicists Congress, June 2003.

2. Z. Salman: VI International Workshop on Applications of Lasers in Atomic and Nuclear Research, Poznan, Poland, 2004.

3. R.F. Kiefl: International Conference on Hyperfine Interactions, Bonn, Germany, 2004.

4. R.F. Kiefl: The Fourth International Conference on Exotic Nuclei and Atomic Masses (ENAM'04), 2004.

5. W.A.MacFarlane: ProRIB 2004: workshop on the production and utilization of radioactive ion beams from isol type facilities, Puri, India, 2004.

6. W.A.MacFarlane: Seaborg Prize Symposium in honor of D.G. Fleming at the American Chemical Society National Meeting, Anaheim, 2004.

7. W.A.MacFarlane: Collaboratory on Electron Correlations - Toward a New Research Network between Physics and Chemistry", Japan, 2005.

0.0.9 Theses etc.

Undergraduate Reports:

1. "Neutral Beam Monitor for β-NMR", Eric Reynard, UBC Physics 2003

2. "Preparation and Characterization of Niobium Thin films and Crystals", Jialin Shi, UBC Chemistry, NSERC USRA 2003

3. "Calculation of Dipolar Line Width", Samir Tabbara, TRIUMF Summer Student 2003

4. "Examining the Interlayer Exchange Coupling in an Fe/Ag (001) Multilayer for βNMR", Todd A. Keeler, TRIUMF Summer Student 2003

5. "Time Evolution of the Nuclear Spin Polarization of ^{8}Li$^+$", Mike Smadella, TRIUMF Summer Student, 2004

6. "Analysis of βNMR data for Cu and Ge", Brendan Campbell, U of A Physics, NSERC USRA 2004

7. "Spectroscopic Determination of the Interaction Between Endohedral Lithium and Buckminster Fullerene usind β-Detected NMR", Jahangir Valiani, UBC Chemistry Honours Thesis, 2005

8. "Point Charge Simulations of the Electric Field Gradient", Jordan Daniel Schultz, UBC Physics Honours Thesis, 2005