Experiment 815, 816 and 817: β-NMR

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The positive muon beams at TRIUMF and associated instruments are an excellent way to probe the magnetic properties of bulk materials. Over the past few years we have developing a closely related technique called beta detected NMR which uses the low energy radioactive ion beams from ISAC. These instruments complement muon spin rotation in that they can be used to probe magnetic and electronic properties of nanostructures and ultrathin films. Recently we have commissioned a second beta-NMR spectrometer for carrying out nuclear quadrupole resonance in zero static external field. In particular we have observed nuclear quadrupole resonances of ^{8}Li in several crystals such as $SrTiO_3$, Al_2O_3 and Sr_2RuO_4. We anticipate that, as in the case of μSR, the ability to carry out measurements in zero external magnetic field will have significant applications in the area of magnetism and superconductivity. More details are given in the next section.

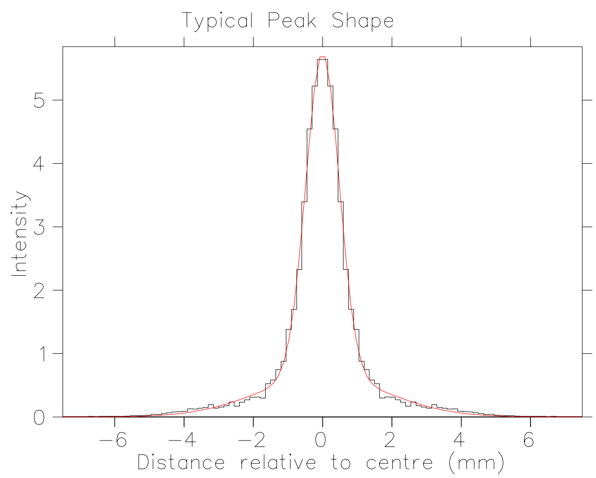

Fig. 1. A quantitative measure of the beamspot as measured by a CCD camera imaging a scintillator mounted at the sample position.

We also made progress in quantifying the beamspot imaging that we perform with a CCD camera by correlating these images with the β count rates. An example of the light output in a section through the beamspot is shown in Fig. 1, indicating the high quality of the beamspot for this energy and magnetic field.

0.0.1 β-Detected Pure Quadrupole Resonance

The most significant advance over the last year is the demonstration of β-detected nuclear quadrupole resonance. Nuclei with spins greater than $\frac{1}{2}$ possess electric quadrupole moments which couple the nuclear spin to the local gradient of the electric field (EFG). In a single crystal for a single site with an axially symmetric EFG, this leads to an effective spin hamiltonian of the form[1]: $H_Q = \frac{1}{6}h\nu_Q[3m^2 - I(I+1)]\delta_{mm'}$, where $h\nu_Q$ is the scale of the quadrupolar coupling, I and m are the nuclear spin quantum number and the magnetic quantum number with a quantization axis (z) defined by the direction of maximal EFG. The quadrupolar spectrum is thus very simple with just two transition frequencies at $\frac{\nu_Q}{2}$ and $\frac{3\nu_Q}{2}$. Provided the initial polarization has a component in the z direction, then we should observe a resonant loss of the β decay asymmetry when the frequency of H_1 (the oscillating transverse magnetic field) matches the quadrupolar level splitting.

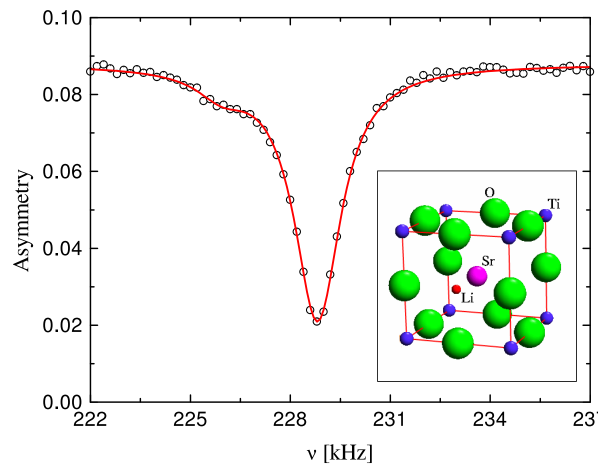

Fig. 2. The β-NQR signal of interstitial ^{8}Li $SrTiO_3$. The spectrum is taken at room temperature in zero applied field.

By analogy with conventional nuclear magnetic resonance, we call this zero field technique βNQR. For light nuclei like ^{8}Li, the nuclear electric quadrupole moment is small (+32 mB), so the quadrupole coupling ν_Q is small. For ^{8}Li we expect it to be less than 200 kHz in most cases. These low frequencies were beyond the range of the RF system designed for the high field spectrometer. Consequently we developed a radio/audio frequency irradiation system which can irradiate either with a single frequency ν or an equal combination of ν and 3ν. In the case of high initial polarization, only the $m = \pm 2$ state is initially occupied and irradiation at $\frac{3\nu_Q}{2}$ will yield a small change in the asymmetry as the transition is saturated. To achieve full saturation, i.e. equalization of the populations of the magnetic states, yielding zero asymmetry, we thus require the two frequency irradiation. Fig. 2 shows such a signal in an epitaxially prepared $\langle 100 \rangle$ crystal of $SrTiO_3$ at room

[1] see "Nuclear Quadrupole Resonance Spectroscopy" by T.P. Das and E.L. Hahn (Academic, NY, 1958) for example.

temperature. We expected this value for ν_Q from our prior observations of quadrupolar splitting of the high field resonance in this material.

0.0.2 Conventional Metals

Previously we observed a split resonance of ^{8}Li near the Larmor frequency in high magnetic field (3 Tesla) in Ag and Au. We have proposed that this was due to 2 distinct Li sites differing in their Knight shifts. In order to test this hypothesis it was necessary to carry out measurements in much lower magnetic field (0.3T) where the back detector has a very small effective solid angle due the the reduced magnetic field. This is now possible since we have also commissioned a new *Neutral Beam Monitor* (NBM), which is detects βs from polarized neutral Li that is not reionized in the He cell of the polarizer. This in turn allows measurements with a single forward detector which can be normalised to the incoming beam rate as measured at the NBM. The NBM is now commissioned and fully integrated into the data acquisition system. The data on Ag were taken at 0.3 T and show a very narrow line at all temperatures. This confirms that the lines at high field are split because of a site dependent Knight shift.

In Ag and Au we have observed the split resonance now in several samples, but in addition only in Au in a temperature range around 225 K, at 0.3 T, we find a broad line in addition to the narrow one corresponding to the 2 cubic sites (unresolved here). Fig. 3 shows an example of this line. We speculate that it is due to a transition state, i.e. the Li spends significant time just adjacent to a local-cubic-symmetry-breaking vacancy. It occurs in the same temperature regime where the amplitudes of the two lines we resolve at high field are changing.

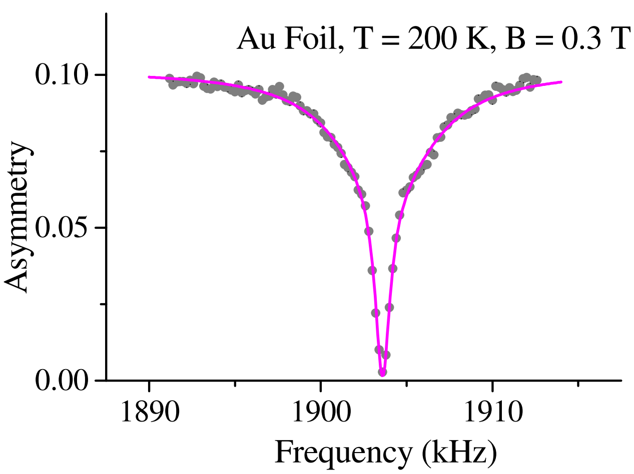

Fig. 3. The βNMR signal of interstitial ^{8}Li in a high purity annealed Au foil. This data was taken with 1.7 watts of RF power and full beam energy (30 keV). Notice the broad line superimposed on the narrow line at the larmor frequency which is likely due to Li that is trapped adjacent to an Au vacancy.

[2] C. Prigge *et al.*, Z. Phys. Chem. **189**, 153 (95).

Having developed the low frequency capability in order to measure the βNQR, we also demonstrated that we can observe conventional βNMR resonances in very small applied magnetic fields. Note the βNQR spectrometer at the polarimeter station is equipped with a Helmholtz coil allowing a static field of up to 0.015 T to be applied. Fig. 4 shows such a resonance. Conventional NMR relies on thermal polarization of the nuclear spins (unlike the highly athermal polarization produced optically in the ^{8}Li beam), thus resonance in such low fields is nearly impossible.

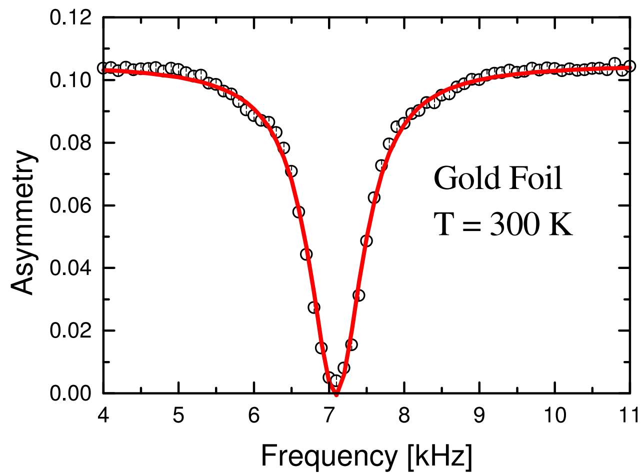

Fig. 4. The β-NMR signal of interstitial ^{8}Li in annealed high purity gold at room temperature in a field of about 11 Gauss.

0.0.3 Superconductors

Recently, we have implanted ^{8}Li near the surface of a conventional superconductor $NbSe_2$. We find a remarkably small quadrupolar frequency ν_Q for interstitial ^{8}Li, even though the crystal is noncubic. ¿From the asymmetry of the resonances for the two nuclear helicities, we estimate that $\nu_Q < 1$ kHz, considerably smaller than what is seen in analogous stable Li intercalation compounds[2]. Above the critical temperature we observe a single resonance at the Larmor frequency with unresolved quadrupolar effects.

When a type II superconductor is cooled in a magnetic field H, for H in a certain range, the field will become inhomogeneous due to the formation of a lattice of magnetic vortices, a consequence of the superconductors tendency to screen the field. Such a field inhomogeneity gives rise to the well-known broadening of the magnetic resonance in the superconducting state. In Fig. 5, we detect this effect by observing a broadened line possessing the expected long high frequency tail. Establishing the technique in a well studied superconductor such as $NbSe_2$ is an important step towards the application of βNMR to the study properties of the vortex state in exotic and thin film superconductors.

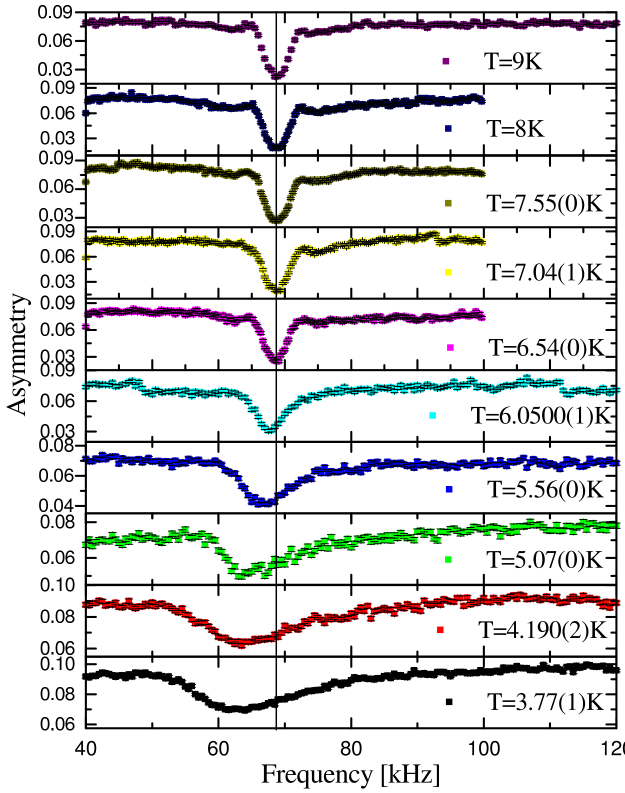

Fig. 5. The β-NMR signal of interstitial ^{8}Li in a crystal of $NbSe_2$ with $H//c$ (the hexagonal axis) The field H was about 110 G.

0.0.4 Semiconductors

We have also begun experiments on semiconductors in which the implanted radioactive atom is used to simulate the behaviour an isolated Li impurity. The inset of Fig. 6 shows the βNMR resonances at room temperature and at low tmeperature. In both cases there is a large amplitude line which is attributed to Li at sites which are close to being cubic and thus have no resolved quadurupole splitting. There is however a strong temperature dependence to the line width shown in main part of Fig. 6. At low temperatures the width is attributed to nuclear dipolar broadening from the abundant ^{69}Ga, ^{71}Ga and ^{75}As nuclear moments. A slight decrease as the temperature increases from 5K to 150 K is attributed to motional narrowing as the Li begins to hop. However, above 150 higher temperatures the line broadens significantly. This may be to small unresolved quadrupolar splittings as the Li approaches the vacancies which are presumably created when the Li is implanted. Note the line width peaks at about 290K. The drop at higher temperatures is likely due to the Li occupying the vacancy where the quadrupolar splitting is identically zero. This behaviour is very similar to that seen in the annealed Au foil. Further measurements are required to confirm this scenario.

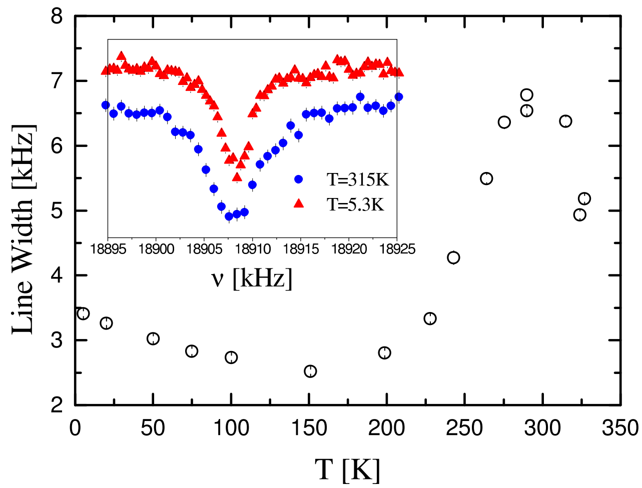

Fig. 6. The linewidth of the β-NMR resonance of ^{8}Li in semi-insulating high purity GaAs with $H \parallel < 100 >$.

0.0.5 Publications

"Quadrupolar Split ^{8}Li βNMR in $SrTiO_3$", W.A. MacFarlane, G.D. Morris, K.H. Chow, R.A. Baartman, S. Daviel, S.R. Dunsiger, A. Hatakeyama, S.R. Kreitzman, C.D.P. Levy, R.I. Miller, K.M. Nichol, R. Poutissou, E. Dumont, L.H. Greene, R.F. Kiefl, Proceedings of the 9^{th} International Conference on Muon Spin Rotation, Physica B **326**, 209-212 (2003).

"^{8}Li βNMR in Thin Metal Films", W.A. MacFarlane, G.D. Morris, T.R. Beals, K.H. Chow, R.A. Baartman, S. Daviel, S.R. Dunsiger, A. Hatakeyama, S.R. Kreitzman, C.D.P. Levy, R.I. Miller, K.M. Nichol, R. Poutissou, R.F. Kiefl, Proceedings of the 9^{th} International Conference on Muon Spin Rotation, Physica B **326**, 213-216 (2003).

"Range Straggling of Low Energy ^{8}Li$^+$ in Thin Metallic Films using βNMR" T.R. Beals, R.F. Kiefl, W.A. MacFarlane, K.M. Nichol, G.D. Morris, C.D.P. Levy, S.R. Kreitzman, R. Poutissou, S. Daviel, R.A. Baartman, K.H. Chow, Proceedings of the 9^{th} International Conference on Muon Spin Rotation, Physica B **326**, 205-208 (2003).

"Low Energy Spin Polarized Radioactive Beams as a Nanoscale Probe of Matter: Progress and Perspective", R.F. Kiefl, W.A. MacFarlane, G.D. Morris, P. Amaudruz, D. Arsenau, H. Azumi, R. Baartman, T.R.

Beals, J. Behr, C. Bommas, J.H. Brewer, K.H. Chow, E. Dumont, S.R. Dunsiger, S. Daviel, L. Greene, A. Hatakeyama, R.H. Heffner, Y. Hirayama, B. Hitti, S.R. Kreitzman, C.D.P Levy, R.I. Miller, M. Olivo and R. Poutissou, Proceedings of the 9^{th} International Conference on Muon Spin Rotation, Physica B **326**, 189-195 (2003).

"Low Energy Spin Polarized Radioactive Beams as a Probe of Thin Films and Interfaces", R.F. Kiefl, W.A.MacFarlane, P. Amaudruz, D. Arsenau, R. Baartman, T.R. Beals, J. Behr, J. Brewer, S. Daviel, A. Hatakeyama, B. Hitti, S.R. Kreitzman, C.D.P Levy, R. Miller, M. Olivo, R. Poutissou, G.D. Morris, S.R. Dunsiger, R. Heffner, K.H. Chow, Y. Hirayama, H. Azumi, C. Bommas, E. Dumont and L.H. Greene, Nuclear Instruments and Methods in Physics Research B, **204**, 682-688 (2003).

0.0.6 Theses etc.

Undergraduate Reports:

"Preparation and Characterisation of Thin Films for βNMR" by K.M. Nichol, UBC

"Dipolar Linewidth Calculations and Spin Diffusion" by S.X.Y. Wang, UBC

Appendix D and Appendix E update:

Updated Collaboration List:

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