Beta-Detected NQR in Zero Field with a Low Energy Beam of $^8$Li$^+$

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Abstract

Beta-detected nuclear quadrupole resonances (β-NQR) at zero field are observed using a beam of low energy highly polarized radioactive $^8$Li$^+$. The resonances were detected in SrTiO$_3$, Al$_2$O$_3$ and Sr$_2$RuO$_4$ single crystals by monitoring the beta-decay anisotropy as a function of a small audio frequency magnetic field. The resonances show clearly that $^8$Li occupies one site with non-cubic symmetry in SrTiO$_3$, two in Al$_2$O$_3$ and three sites in Sr$_2$RuO$_4$. The resonance amplitude and width are surprisingly large compared to the values expected from transitions between the $|\pm 2\rangle \leftrightarrow |\pm 1\rangle$ spin states, indicating a significant mixing between the $|\pm m\rangle$ quadrupolar split levels.

Key words: Zero field, quadrupole resonance, β-NQR, β-NMR.

1. Introduction

Recently, we have constructed two spectrometers for beta-detected nuclear magnetic resonance [1] (β-NMR) and nuclear quadrupole resonance [2] (β-NQR), using a low energy highly polarized $^8$Li$^+$ beam. We report here on the first results obtained from the β-NQR spectrometer at zero applied magnetic field. The β-NQR spectra were obtained for $^8$Li implanted into single crystals of SrTiO$_3$, Al$_2$O$_3$ and Sr$_2$RuO$_4$. The ability to perform measurements in zero applied field has many potential applications in studies of magnetism and superconductivity. It is also remarkable that the resonances are narrow (few kHz) and easily observed at acoustic frequencies.

This work is intended as an initial characterization of the spectra of $^8$Li in SrTiO$_3$, Al$_2$O$_3$, and Sr$_2$RuO$_4$. SrTiO$_3$ is probably the best-studied perovskite transition metal oxide. It is interesting for its prototypical soft mode structural phase transition (~105 K) [3], its ferroelectric properties [4,5], and as a high dielectric constant layer in heterostructures based on Si [6]. Both SrTiO$_3$ and Al$_2$O$_3$ are important substrate materials for thin films, and therefore it is important to understand the behavior of $^8$Li in these substrates for future studies. Sr$_2$RuO$_4$ is an unconventional spin-triplet superconductor ($T_c = 1.5$ K) as demonstrated by
NMR Knight shift measurements [7]. In its normal state it also exhibits a highly correlated metallic behavior which can be described as a quasi-two-dimensional Fermi liquid [8,9].

The experiment was performed at the TRIUMF ISAC facility in the new $\beta$-NQR spectrometer. In this experiment a highly polarized beam of $^{8}\text{Li}$ is implanted in the sample with energy of 30 keV. At this energy the average implantation depth is $\sim 200$ nm. A linearly polarized oscillating magnetic field $B_1$ is applied perpendicular to the initial nuclear spin polarization. A resonant loss of the nuclear beta decay asymmetry of $^{8}\text{Li}$ occurs when the frequency of the oscillating field $\nu$ matches the nuclear energy spin level splitting. A more detailed description of the $\beta$-NQR spectrometer and of the beta detected nuclear resonance technique used can be found in Ref.[2] and references therein. Once the development of this spectrometer is completed it will have the capability to reduce the beam energy to 100 eV, allowing for depth profiling measurements on a nm scale in zero applied magnetic field.

The Hamiltonian for the implanted $^{8}\text{Li}$ at zero field in the presence of an axially symmetric electric field gradient (EFG) is [10]

$$H_q = \hbar \nu_q [I_z^2 - 2]$$

(1)

where $\nu_q = e^2 qQ/8$, $eq = V_{zz}$ is the electric field gradient at the $^{8}\text{Li}$ site, and $Q$ is the electric quadrupole moment of the nucleus. Therefore, when $q \neq 0$ even at zero magnetic field a splitting between the nuclear spin sub-levels $|m\rangle$ is present (see Fig. 1). In this case two resonance frequencies are possible, one at $\nu = \nu_q$ due to the transitions $| \pm 1 \rangle \leftrightarrow |0\rangle$, and another at $\nu = 3\nu_q$ due to $| \pm 2 \rangle \leftrightarrow | \pm 1 \rangle$ transitions.

2. Results

$\beta$-NQR spectra were collected on an epitaxially polished (100) single crystal of SrTiO$_3$ (Applied Crystal Technologies). We have established earlier that the $^{8}\text{Li}$ occupies the face centered cite in SrTiO$_3$ [2,11], and therefore experiences a non-vanishing EFG. Indeed a large and sharp NQR resonance was observed at $3\nu_q = 228.8(2)$ kHz with width 1.7 kHz corresponding to the $| \pm 2 \rangle \leftrightarrow | \pm 1 \rangle$ transitions (see Fig. 2), whereas the resonance near $\nu_q$ was barely visible, indicating very low probability of $^{8}\text{Li}$ in the $| \pm 1 \rangle$ spin states as expected from the high polarization of the $^{8}\text{Li}$ beam. Note the resonance in Fig. 2 is slightly asymmetric. The solid curve is a fit assuming two overlapping lines. The smaller amplitude line occurs at a slightly lower frequency $\nu = 225.9(2)$ kHz and has a width of about 1.9(1) kHz which is similar to the higher frequency line at $\nu = 228.8$ kHz.

In Al$_2$O$_3$ (epitaxially polished, Honeywell) and Sr$_2$RuO$_4$ (freshly cleaved) the beta decay asymmetry was found to be zero when the $^{8}\text{Li}$ was implanted with its nuclear polarization perpendicular to the c-axis. However, when the sample was rotated by 45° significant asymmetry was

![Nuclear Polarization vs Frequency](image-url)

Fig. 2. The $\beta$-NQR spectrum in SrTiO$_3$ at room temperature and zero applied field.
observed, a clear indication that the EFG experienced by the implanted $^8$Li is along the c-axis in these crystals. In Al$_2$O$_3$ the spectrum (see Fig. 3) shows two distinct resonance lines at $\nu = 92.94(8)$ and $188.9(3)$ kHz, with widths of $15.1(3)$ and $22.5(9)$ kHz respectively. The two resonances are due to at least two inequivalent $^8$Li sites. However, note that the widths of the lines in Al$_2$O$_3$ are significantly larger than that observed in SrTiO$_3$, likely due to a larger distribution of $\nu_q$, which may be caused by multiple $^8$Li sites with very similar values of $\nu_q$. This may be expected considering the complexity of the lattice structure of Al$_2$O$_3$.

The $\beta$-NQR spectrum in Sr$_2$RuO$_4$ is shown in Fig. 4 at room temperature and zero field, where three sharp and separated resonances are observed at $\nu = 7.72(4), 11.57(1)$ and $15.51(2)$ kHz, with corresponding width of $0.55(13), 1.28(3)$ and $0.83(7)$ kHz, indicating three well defined $^8$Li lattice sites in this tetragonal material. The resonances here are even sharper than in SrTiO$_3$, reflecting the high quality of this crystal.

In all three cases presented, the amplitude of the resonances are much larger than expected. The optical pumping method used to generate the nuclear polarization of the $^8$Li beam produces polarization as high as $\sim 70\%$ along the measurement axis ($z$). This polarization can be defined as

$$P_z = \frac{1}{2} \sum_{m=-2}^{m=+2} p_m m$$

(2)

where $p_m$ is the probability that the $|m\rangle$ state is occupied. Realistic values for these probabilities are

$\{p_{+2}, p_{+1}, \cdots, p_{-2}\} = \{0.65, 0.2, 0.15, 0, 0\}$.

When the frequency of $B_1$ is $\nu = 3\nu_q$, and assuming sufficient power to saturate the transition, the probability that the $|+2\rangle$ or $|+1\rangle$ are occupied becomes equal. This implies that the polarization is reduced from its initial value, 0.75, to 0.6375 with probabilities $\{0.425, 0.425, 0.15, 0, 0\}$, i.e. 15% loss in the polarization or asymmetry. Our measurements show a considerably larger amplitude. This enhancement can be explained by an additional term in the Hamiltonian, which mixes the different sub-levels $|m\rangle$, and produces a larger effect by allowing transitions other than $|+2\rangle \leftrightarrow |+1\rangle$. Such a term can be produced by a small stray magnetic field perpendicular to the EFG axis, or by non-axial terms in the EFG. In the case of SrTiO$_3$ we concluded that the non-axial terms in the EFG are responsible for this enhancement [2]. However, more measurements are required on Al$_2$O$_3$ and Sr$_2$RuO$_4$ to identify the origin of the additional interaction responsible for the amplitude enhancement.

Interestingly, in addition to the large amplitudes, the sum of the amplitudes in the case of
Al$_2$O$_3$ and Sr$_2$RuO$_4$ is larger than 1, as seen in Fig. 3 and 4. This is only possible if $^8$Li is diffusing between inequivalent sites. When the diffusion rate is higher than the $^8$Li decay rate, it would be possible for a $^8$Li particle which is initially off resonance to move to an on resonance site, thus increasing the measured amplitude. This will be verified by cooling the sample and measuring the $\beta$-NQR spectra as a function of temperature, where one expects the diffusion rate to slow down, and consequently the sum of amplitudes to drop below 1.

3. Summary and Conclusion

We have demonstrated that it is possible to carry out $\beta$ detected nuclear quadrupole resonance using a beam of low energy highly polarized $^8$Li$^+$. Clear $\beta$-NQRs were observed in SrTiO$_3$, Al$_2$O$_3$, and Sr$_2$RuO$_4$ indicating that the implanted Li adopts well defined crystalline lattice sites. In contrast to $\mu^+$, the quadrupole resonances provide a means of identifying the $^8$Li site.

There is evidence for small terms in spin Hamiltonian which lead to mixing of the $| \pm m \rangle$ states and a dramatic enhancement of the amplitude of the resonances at $3\nu_q$. The ability to perform $\mu$SR in zero field has been used extensively in studies of magnetism and and superconductors. We anticipate similar applications are possible with $\beta$-NQR in studies of ultra-thin films and interfaces. For example in superconductors it could be used to measure the absolute value of the London penetration depth or to search for states with broken time reversal symmetry. In semiconductors or ionic compounds it can be used to study the diffusion and electronic structure of isolated Li in reduced geometries.

Acknowledgements This work was supported by the CIAR, NSERC and TRIUMF. We thank Rahim Abasalti and Bassam Hitti for technical support. We also thank Laura Greene for providing the SrTiO$_3$ sample.

References

[1] G. D. Morris W. A. MacFarlane K. H. Chow Z. Salman D. J. Arseneau S. Daviel A. Hatakeyama S. R. Kreitman C. D. P. Levy R. Poutissou R. H. Heffner J. E. Elenewski L. H. Greene and R. F. Kiefl. Phys. Rev. Lett., 93:157601, 2004.

[2] Z. Salman E.P. Reynard W.A. MacFarlane K.H. Chow J. Chakhalian S.R. Kreitman S. Daviel C.D.P. Levy R. Poutissou and R.F. Kiefl. Phys. Rev. B, 70:104404, 2004.

[3] R.A. Cowley. Phil. Trans. R. Soc. London A, 354:2799, 1996.

[4] J.G. Bednorz and K.A. Müller. Phys. Rev. Lett., 52:2289, 1984.

[5] M. Itoh R. Wang Y. Inaguma T. Yamaguchi Y.J. Shan and T. Nakamura. Phys. Rev. Lett., 82:3540, 1999.

[6] R.A. McKee F.J. Walker and M.F. Chisholm. Phys. Rev. Lett., 81:3014, 1998.

[7] K. Ishida H. Mukuda Y. Kitaoa K. Asayama Z.Q. Mao Y. Mori and Y. Maeno. Nature, 396:658, 1998.

[8] A.P. Mackenzie S.R. Julian A.J. Diver G.J. McMullan M.P. Ray G.G. Lonzarich Y. Maeno S. Nishizaki and T. Fujita. Phys. Rev. Lett., 76:3786, 1996.

[9] Y. Maeno et. al. J. Phys. Soc. Jpn., 66:1405, 1997.

[10] C.P. Slichter. Principles of Magnetic Resonance. Springer-Verlag, New York, third edition, 1990.

[11] W. A. MacFarlane G. D. Morris K. H. Chow R. A. Baartman S. Daviel S. R. Dunsiger A. Hatakeyama S. R. Kreitman C. D. P. Levy R. I. Miller K. M. Nichol R. Poutissou E. Dumont L. H. Greene and R. F. Kiefl. Physica B, 326:209, 2003.