Range straggling of low energy ${}^{8}Li^{+}$ in thin metallic films using β -NMR

<u>T.R. Beals</u>^a, R.F. Kiefl^{a,c,d}, W.A. MacFarlane^{b,d}, K.M. Nichol^{a,d}, G.D. Morris^e, C.D.P. Levy^d, S.R. Kreitzman^d, R. Poutissou^d, S. Daviel^d, R.A. Baartman^d and K.H. Chow^f

^aDepartment of Physics and Astronomy, University of British Columbia, Vancouver, BC, Canada

^bChemistry Department, University of British Columbia, Vancouver, BC, Canada ^cCanadian Institute for Advanced Research

^dTRIUMF, 4004 Wesbrook Mall, Vancouver, BC, V6T 2A3, Canada

^eLos Alamos National Laboratory, MS K764, Los Alamos, NM 87545 USA

^fDepartment of Physics, University of Alberta, Edmonton, AB, Canada

Abstract

Recently it has been demonstrated that low energy spin polarized radioactive nuclei can be used to probe the electronic and magnetic properties of ultra-thin samples using β detected NMR. However, use of the technique requires an accurate understanding of ion penetration depth and range straggling as a function of beam implantation energy and target density. Range straggling measurements of ⁸Li were conducted with thin films of metallic Al and Au on sapphire and SrTiO₃ substrates and the results were compared to SRIM code predictions.

Key words: β -NMR, range straggling, thin metallic films

Corresponding author:

T.R. Beals, Department of Physics & Astronomy, University of British Columbia, Vancouver, BC V6T 1Z1, Canada. Tel. 604-839-0379, FAX: 604-822-5324, e-mail: trbeals@physics.ubc.ca

In order to fully utilize low energy spin polarized radioactive ions as a probe of interfaces and thin films [1], it is necessary to establish the stopping distribution in typical solids, since this information is needed to interpret any depth sensitive measurement. Numerical Monte Carlo programmes exist to calculate such curves (e.g. SRIM [2]), but they are largely untested at the low energies



Fig. 1. Spectra in 50 nm Au film on a SrTiO_3 substrate, in a 3.0 T magnetic field. Beam energy is 10 KeV. Both spectra are fitted to a Lorentzian. The two spectra correspond to "up" and "down" helicities. A is the resonance amplitude, while $2A_0$ is the total asymmetry off resonance.

required for near-surface implantation. To experimentally establish the range curves, we studied the amplitude of the β -NMR resonance in thin metal films deposited on crystalline substrates as a function of incident ion energy. This is similar to what has been done with low energy muons [3]. At room temperature, FCC metal films produce simple resonances [4], which are quite distinct from the substrate resonances [5]. In the case of sapphire, no resonance of ⁸Li has been observed. One important aspect of the implantation of low energy ions is backscattering; this leads to ions stopping outside the sample region. In the Rutherford model, the cross-section for backscattering goes as the square of the nuclear charge Z, so we expect large differences between the Al and Au when backscattering is important.

The 50 nm Au sample was electron beam evaporated on a $\mathrm{SrTiO}_3 \langle 100 \rangle$ substrate and had a surface roughness of about 0.45 nm as measured with atomic force microscopy [4]. The 150 nm Al sample was grown on epitaxially-polished c-axis $\langle 0001 \rangle$ Al₂O₃ substrate, and had a surface roughness about an order of magnitude greater than the Au film (4.5 nm). Each of the samples was clamped to an Al sample holder held at room temperature. The incoming 30.5 KeV ⁸Li⁺ beam was longitudinally polarized in flight using an optical pumping scheme [1]. The spectrometer sat on a high voltage platform whose voltage allowed



Fig. 2. Spectra in 150 nm Al film on an Al₂O₃ substrate, in a 3.0 T magnetic field. Beam energy is 15 KeV. Both spectra are fitted to a modified exponential. The two spectra correspond to "up" and "down" helicities. A is the resonance amplitude, while $2A_0$ is the total asymmetry off resonance.

the implantation energy to be adjusted. In addition, the nuclear polarization could be flipped by reversing the laser helicity. β –NMR spectra were taken with both helicities so that the resonance amplitude could be normalized to the full β decay asymmetry. To tune the beam optics, a thin scintillator was placed at the sample position and viewed with a CCD camera. For most energies the beamspot (~ 3 mm diameter) was considerably smaller than the beam access window of the cryostat (8 mm by 8 mm). However, the beamspot images grew in size and became less intense at energies below 5 KeV. All the resonances were measured in a magnetic field of 3 T applied along the axis of the beam. In addition, there was a small RF magnetic field of 0.1 mT applied perpendicular to the main field whose frequency was swept slowly through the resonance. On resonance, the RF field was sufficient to destroy all the nuclear polarization, i.e. saturate the main part of the resonance.

Fig. 1 shows a typical β -NMR resonance in the Au film for each of the two beam helicities. The curves are fits to a Lorentzian, as expected for a fully saturated line. Without backscattering, the full asymmetry off resonance (A_0) is proportional to the total Li in the metal film plus substrate and is determined by the properties of the nuclear decay, the geometry of the detectors, and the nuclear spin polarization. The ratio between the resonance amplitude A and



Fig. 3. Total asymmetry $(2A_0)$ versus beam energy, for the 50 nm Au sample (circles) and the 150 nm Al sample (filled triangles). Note the drop in total asymmetry at lower beam energies.

 A_0 is then a measure of the fraction of ⁸Li in the metal film. Surprisingly, the spectra in the Al film were not simple Lorentzians, but exhibited an unusual flat-topped lineshape, which fit well to a form $Ae^{-|(\nu-\nu_0)/\Delta|^{\alpha}}$, where $\alpha \approx 5$ and $\Delta \approx 33$ KHz. Example spectra are shown in Fig. 2. Nevertheless, the spectra appeared to be saturated even though they were considerably broader due to the large nuclear dipole moment of ²⁷Al.

Fig. 3 shows $A_0(E)$ as a function of energy for both the Au and Al films. Note that the $A_0(E)$ is slightly larger in Al than in Au for E above 5 KeV. This is consistent with greater backscattering from the Au film. Surprisingly, $A_0(E)$ drops sharply for implantation energies below 5 KeV in both the Au and Al films. This is unlikely to be a consequence of backscattering, since it is the same for Al and Au. The drop-off is instead attributed to a change in the beam optics since the decrease appears in the same energy range where the beam focusing deteriorated.

As mentioned above, the ratio A/A_0 is a measure of the fraction of ions stopping in the film. This quantity, averaged for the two helicities, is plotted in Fig. 4 for both the Au and Al films. The measurement in Au agrees with the SRIM calculations at higher energies but $A/A_0(E)$ does not extrapolate



Fig. 4. Top: Normalized amplitude (A/A_0) versus beam energy for the 50 nm Au sample (circles). Bottom: Normalized amplitude (A/A_0) versus beam energy for the 150 nm Al sample (filled triangles). SRIM predictions are shown for comparison.

to the expected value of 1.0 in the low energy limit. This implies there is a fraction of the Li which does not contribute to the resonance, possibly due to backscattering since the missing fraction is close to what SRIM predicts. Although the linewidth in the gold film is slightly broader than in high purity foil[1] the main part of the line should still be saturated.

The ratio $A/A_0(E)$ in Al for energies above 5 KeV looks more reasonable since it appears to be approaching 1.0 at zero energy. However there is a sudden downturn in A/A_0 at lower energies, which is not understood. Since backscattering is predicted to be negligible for Al, the most plausible explanation is incomplete saturation of the line at low energies. For example, a broader component might arise from surfaces and grain boundaries. There is evidence for such a broad component in the ¹²B β –NMR data in Pt[6]. Since the linewidth is broader in the Al due to the larger nuclear moments of ²⁷Al, the resonance amplitude in the Al film may be more sensitive to any increased line broadening associated with the surface. One would expect such additional line broadening near any metal surface due to variations in the electron density which are analogous to Friedel oscillations around a pointlike charged impurity. These Bardeen–Friedel oscillations decay away from the surface and act to broaden the conventional NMR corresponding to nuclei near metalllic surfaces (see [4]).

In slow muon ranging experiments, there is a significant reduction of the metal film signal (equivalent to A/A_0) at low energies, which has been attributed to backscattering effects [3]. The effect we see in Al is somewhat different in appearance (note the scatter in A/A_0 at 2 and 3 KeV), but might be of the same nature as that observed with slow muons.

In conclusion, we have carried out β -NMR measurements on thin films of Au and Al as a function of implantation energy. The Au data agrees reasonably well with SRIM predictions at high energies, but the agreement for Al is poor. Further measurements are required to determine the influence of backscattering and surface broadening.

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