Experiment 1040 β -NMR Investigation of the Magnetic Properties of Manganites

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The manganites exhibit a range of interesting behaviour which includes large magneto-resistance, a sharp insulator-metal transition and a rich magnetic phase diagram. Numerous probes have shown that the magnetic transitions are unconventional, involving both phase separation and phase competition. In $La_{1-x}Ca_{x}MnO_{3}$ (LCMO) (x > 0.2), it is proposed that a strong coupling of the charge and spin degrees of freedom arises from double exchange and strong Hund coupling between the spins of the itinerant Mn charge carriers and the core spins. In materials with low doping (x < 0.2), Jahn-Teller effects and polarons dominate the behaviour. In many of these materials, charge order, orbital order, ferromagnetic, antiferromagnetic and paramagnetic phases can be induced with small changes in temperature, an external magnetic field, chemical composition, and light levels. Given this one would expect their properties to change as a function of distance from a surface or interface. Although many experimental methods have been applied to these materials, including μ SR, neutron scattering, and NMR, none can be used to probe local properties in a depthcontrolled manner.

In this experiment we are using low energy β -NMR to monitor magnetic properties of a thin film of La_{1-x}Ca_xMnO₃ with x = 0.30. The main result is the implanted ⁸Li's spin relaxation rate is sensitive to the LCMO magnetic fluctuations both in the film and in the substrate. This shows that low energy β -NMR can be used to probe spin dynamics in thin films and near the surface of a material as a function of depth.

The LCMO film was synthesised by pulsed laser deposition at the University of Toronto on epitaxially polished SrTiO₃ substrate and has a thickness of approximately 2,500Å. The film exhibits a sharp insulatormetal transition at 260K, such that it is semiconducting above T_c and metallic at lower temperatures. The magnetic properties also change at T_c since it is ferromagnetic below T_c . Although the measurements presented here were performed at a single implantation energy, a high voltage platform is being constructed for the low field spectrometer. This will allow the mean depth of implantation to be controlled on a nm lengthscale. Fig. 1 shows the simulated implantation profile for a 28 keV ⁸Li ion, the beam energy used in this experiment. The peak in the distribution occurs near the middle of the film $(1,500\text{\AA})$ such that only about 5% of the beam lands in the substrate.

Fig. 2 shows the spin relaxation measurements in

a small magnetic field (B = 150 G) at a few different temperatures. The measurements were taken by delivering relatively long (4s) beam pulses separated by long periods (12s) of beam off. The beta-decay asymetry is measured during the beam-on and beam-off periods, although only the beam-on period is shown in Fig. 2. In order to fit the data one must convolute the theoretical relaxation function with the square beam pulse. Typically a short beam pulse would be used for this purpose, in which case no convolution is required. However, we have found that the statistics and resulting signal quality are much higher with the long pulse method. At low temperature (Fig. 2a [T=10K]), the initial asymmetry is close to the full asymmetry expected (0.19) and most of the relaxation is visible in the ⁸Li time window. A fast component relaxes on a timescale of 100ms while most of the signal relaxes on the timescale of seconds. As the temperature increases the relaxation rates increase rapidly. Near T_c , the relaxation is so fast the signal is barely visible in our time window (Figs. 2c [T=260K] and 2d [T=290K]). On a long time scale there is still a small amplitude slowly relaxing component, which we attribute to 8 Li in the STO substrate expected from the predicted implantation profile (see Fig. 1.)

Below T_c the spin relaxation rate in STO is about an order of magnitude larger than in a bare STO crystal and about a factor of two larger above T_c . The difference can be attributed to the spin fluctuations in the LCMO film. The ⁸Li in the STO lies mostly within a few hundred Angstroms of the LCMO interface (see Fig. 1) and therefore will experience fluctuating dipolar magnetic fields from the LCMO. This is confirmed by the fact that at T = 260 K, the relaxation rate in the STO increases sharply and is too fast to be seen. These results suggest it is possible to use the signal in the substrate to monitor fluctuations in the film. This could be useful in situations where the relaxation in the film itself is too fast to be observed directly $(T_1 << \tau = 1.2s, \text{ the } ^8\text{Li} \text{ lifetime}).$

Fig. 3 shows the relaxation rate of the fast component, that we attribute to the LCMO film. Near T_c the relaxation rate is outside the ⁸Li time window. Therefore we plot a lower bound for the relaxation rate. Above T_c the signal falls back into our time window. This behaviour is expected for a ferromagnet since the spin relaxation rate should peak near T_c . Above T_c there is a critical slowing down of the spin fluctuations as one approaches the transition from above, whereas well below T_c the relaxation rate in the film decreases due to the freezing out of the magnetic excitations.

In conclusion, we have demonstrated that low energy β NMR can be used to probe magnetic spin fluctuations in thin magnetic films. The spin fluctuations

result in nuclear spin relaxation of the Li both inside the film and also in the substrate next to the film. This opens up a range of possible new experiments on manganites and other thin magnetic films.



Fig. 1. Simulated implantation profile of ⁸Li ions at 28 keV implantation energy in 2,500Å thick LCMO film on $SrTiO_3$ substrate. The implantation profile is calculated with TRIM Monte Carlo simulation.



Fig. 2. Relaxation of the polarization of ⁸Li decay in the $La_{1-x}Ca_xMnO_3$ (with x = 0.33) film measured with ⁸Li β -NMR at various temperatures. A 4 s pulse of Li beam is delivered onto the film starting at t = 0. The curves are fits to the sum of a stretched exponential ($\beta = 0.5$) and an

exponential function. Note that the y-axis for panels c and d are expanded compared to panels a and b. Insets: Polarization on an expanded time scale. The time axis is

the same for each inset. Panel (a) is at the top.



Fig. 3. Relaxation rate of the fast component from fits in Fig. 2. This relaxation is attributed to the LCMO layer.

Publications:

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