Physica B 165&166 (1990) 793–794
North-Holland

NQR STUDIES OF SCANDIUM METAL AT LOW TEMPERATURES

L. POLLACK, E.N. SMITH, R.E. MIHAILOVICH, J.H. ROSS Jr.§, P. HAKONEN†, E. VAROQUAUX‡, J.M. PARPIA, R.C. RICHARDSON

Materials Science Center, Cornell University, Ithaca, NY 14853, USA

We have performed susceptibility and relaxation time measurements on scandium metal using nuclear quadrupole resonance (NQR) techniques at temperatures down to 100μK.

1. INTRODUCTION

In systems which possess nuclear spins greater than I = 1/2 the nuclei have electric quadrupole moments. When these atoms are in a crystal which does not have cubic symmetry, the coupling of the quadrupole moments of the nucleus to the electric field gradient causes a splitting of the nuclear levels. Since this interaction depends solely on the orientation of the nucleus, in the absence of an applied magnetic field the $\pm\, m_I$ levels are degenerate. Typically these splittings are on the order of 10's of MHz, but for some systems where the asymmetry of the crystalline axes is slight the splittings can be significantly smaller. One such system is scandium metal (Sc). The nuclear spin of Sc is 7/2, thus the effect of the electric field gradient on the nuclei is to split the nuclear levels into four doubly-degenerate levels. The fundamental quadrupolar splitting of scandium is 130kHz and the sign of the quadrupole interaction is such that the ground state is the ± 7/2 state. Thus the splitting between the lowest two states, the ($\pm\, 7/2 \leftrightarrow \pm\, 5/2$) is 390kHz; the spacing between the next two levels ($\pm\, 5/2 \leftrightarrow \pm\, 3/2$) is 260kHz; and the ($\pm\, 3/2 \leftrightarrow \pm\, 1/2$) level spacing is 130kHz. In temperature these splittings are 18 μK, 12 μK and 6 μK respectively. Since this temperature is close to the expected operating range of our system, scandium metal should be interesting to observe as a function of temperature.

2. APPARATUS

In our first attempt at studying this system, ten 0.13 mm thick foils of polycrystalline Sc were clamped to a piece of copper which was firmly attached to the nuclear stage of a 65 mole copper nuclear demagnetization bundle. A single 500 turn pickup coil serves as the excitation and receiver coil. The coil surrounds the foils, but does not come into thermal contact with them. The circuit can be tuned with the addition of a capacitor external to the cryostat, allowing us to look for any of the three transitions. A superconducting shield encloses the experiment to protect the sample from fringe fields. The population differences between various states can be measured using pulsed-NQR techniques (1). Due to the variations in the gradient in the local electric field, the T_2^* of the Sc is on the order of 40 μsec. Since the recovery time of the receiving amplifier is comparable to the T_2^* it is only possible to observe spin-echoes at the "high" temperature end of the experiment (T> 1mK). At lower temperatures, as the magnetization increases, it is possible to observe tails of the free induction decay signals (FIDs) from the Sc nuclei. Other possible sources of broadening of the signal come from the inhomogeneities between different foils, from the possible reduction of the H_1 field at the innermost foils due to shielding from the outermost foils, or from the variation of the tipping pulse within a skin depth for a single foil. To minimize these problems a single Sc foil is currently being used. The thermal contact between this foil and the copper stage is made by melting potassium and "soldering" the scandium to the copper holder.

A second coil which contains several of the Sc foils is mounted near the center of the bundle, inside the superconducting magnet. This coil is not shielded from the demagnetizing field (H_o). It can be used to measure the changes in susceptibility at fixed field, and also to explore the field dependence of the quadrupole transitions. The T_2^* of the signal changes as a function of H_o, and at reasonably low values of H_o, such as the chosen final field present at the end of a demagnetization, it is possible to observe FID signals from this coil which persist into the "high temperature" regime (T $\approx$ 10mK) thus observation of this signal is relatively

---

Present addresses: § Physics Dept., Texas A&M Univ., College Station, TX † Low Temp. Lab Otakaari 3A, SF-02150 Espoo, Finland ‡ Universite de Paris Sud, Orsay, France easy. All values of the temperature are derived from measurements of Pt nuclear susceptibility.

3. RESULTS

Preliminary measurements of the susceptibility of the Sc in the second coil in a small field ($H_o = 300$ Gauss) show that Curie's law is satisfied down to temperatures on the order of 0.1 mK. A typical FID signal, taken at 1mK, is shown in figure 1. We have observed changes in the susceptibility in both the 260kHz and the 390 kHz transitions. We have not been able to observe the 130kHz transition on either coil, which is surprising since its expected signal size is greater than that from either of the other transitions (2).

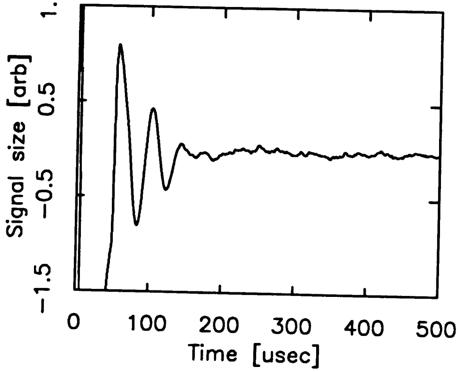

Figure 1 - A typical free induction decay signal

It is also interesting to measure the spin-lattice relaxation times of these systems. NQR systems are expected to show multi-exponential relaxation rates, since the relaxation of each set of levels to equilibrium occurs at an independent rate. For Sc, since there are three sets of levels, a three exponential decay time is expected. The time constants for these rates range from 10 to 60 seconds at 10 mK (3). Measurements made of the return to equilibrium after "saturation" indicate that there are several contributions to the total relaxation, but the shortest time constant we observe is about 10 msec which is significantly shorter than any of the expected spin-lattice relaxation times. If a "fast" process exists that equilizes the spin temperatures of the different levels, the time constant that dominates the equilibration of the system is the shortest spin-lattice relaxation time. A short recovery time would be advantageous in very low temperature experiments. This fast relaxation rate seems to be more or less independent of temperature. It is possibly due to incomplete saturation of the spins since the dimensions of the sample are large compared to the skin depth of the RF. The time constants were measured at the high temperatures by attempting to saturate the spins with the two-pulse sequence that generates the largest echo, then by the examination of a second echo at a variable later time. At lower temperatures smaller pulses were used to minimize heating. The results are displayed in figure 2.

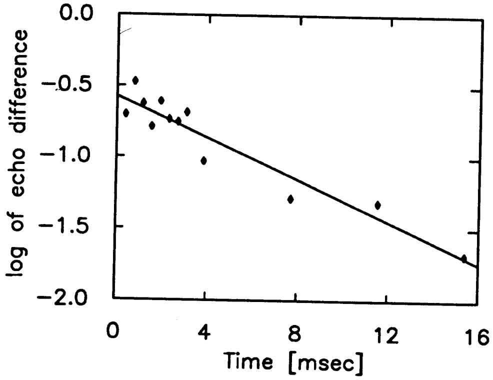

Figure 2 - A relaxation time of 14 msec results from fitting the difference in height of an echo and the "infinite time" echo as a function of the time delay between the saturating sequence and the echo-generating pulses.

4. CONCLUSIONS

Our initial measurements on Sc have yielded some unexpected results. We are currently modifying the system to allow for the detection of FIDs on the magnetically shielded coil. Study of these signals will eliminate questions about heating caused by the 2-pulse echo sequences and should simplify the analysis.

ACKNOWLEDGEMENTS

This work has been supported by the Materials Research Division of the NSF through grant number DMR-8818558. We would like to acknowledge the assistance of: P. Fraenkel, T. Gramila, L. Gunderson and G. Wong. We would also like to thank Professor J. Burlitch for his assistance in preparing the potassium solder joint.

REFERENCES

(1) M. Bloom and R.E. Norberg, Phys. Rev. 93, (1954) 638.
(2) E.I. Fukushima and S.B.W. Roeder, Experimental Pulse NMR (Addison-Wesley, Reading, MA 1981).
(3) A. Narath and T. Fromhold Jr., Phys. Lett. 25A (1967) 49 and M.H. Cohen and F. Reif, Quadrupole Effects in NMR Studies in Solids, in: Solid State Physics, Vol. 5, eds. F. Seitz and D. Turnbull, (Academic Press, NY 1957) pp. 322-448.