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ENHANCED NUCLEAR COOLING AND ANTIFERROMAGNETISM IN HoVO$_4$


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Abstract.- Enhanced nuclear cooling experiments on a single crystal of HoVO$_4$ are reported. The minimum temperature reached was about 1 mK. Antiferromagnetic ordering of the enhanced nuclear spin was observed with a Néel temperature $T_N = 4.5$ mK. A spin-flip transition was observed in an applied magnetic induction of about 140 gauss.

In this paper we report (i) AC susceptibility measurements and (ii) $\gamma$-ray anisotropy measurements following adiabatic demagnetisation.

1. INTRODUCTION.- Holmium vanadate has been investigated by optical spectroscopic and n.m.r. techniques. It is a suitable crystalline material in which to study enhanced nuclear cooling and the ordered state of enhanced nuclear moments. The Ho nuclear relaxation time is of the order seconds at helium temperatures, a large Van-Vleck magnetic moment is extremely short compared to relaxation times in most other non metallic materials. The ionic ground state is a singlet, predominantly $|3/2\rangle$, with a doublet at about 21 cm$^{-1}$ above. As a result HoVO$_4$ exhibits, at helium temperatures, a large Van-Vleck temperature independent susceptibility of 2.32$\mu_B$ per tesla, perpendicular to the tetragonal $c$ axis of the zircon structure. This gives rise to an enhanced nuclear magnetic moment $g_{nu}(1+K)$ where $(1+K)$ is $\approx$ 150 /2.1.

The nuclear hamiltonian may be written, taking the crystal $c$ axis as the $z$ axis,

$$H = -\gamma \hbar /2 \sum_{n=1}^{N} (B \cdot \mathbf{I}) + P \sum_{n=1}^{N} |I(z) - I(\pm 1)|$$

where $P = 25.9$ MHz, $\gamma_1/2\pi = 1526$ MHz T$^{-1}$ and $\gamma_1/2\pi = 20$ MHz T$^{-1}$.

2. EXPERIMENTS.- The single crystals of HoVO$_4$ used in these experiments were grown by the Materials Preparation Group [5] in this laboratory. To obtain a suitably large sample for susceptibility study, two slices cut from a single crystal were cemented together with Araldite to form a cylinder of length 5 mm and diameter 2.5 mm, with the cylindrical axis along the crystal $c$ axis. The radioactive sample was a somewhat smaller crystal, neutron irradiated and subsequently annealed at 1300 K.

Thermal contact to the crystals was made through 200 insulated copper wires soldered to the cold finger of an Oxford Instruments $^3$He/$^4$He dilution refrigerator. The temperature of the cold finger was measured above 50 mK by a carbon resistance thermometer and at lower temperatures by a $^{60}$CoCo nuclear orientation thermometer.

The AC susceptibility was measured using a resonant circuit designed by Robinson [6]. A coil containing the cylindrical sample formed the inductor in a parallel resonant circuit. The resonant frequency is related to the susceptibility $\chi$ by $\omega = \omega_0 f$ where $\omega_0$ is the resonant frequency without the sample, of order 400 kHz and $f$ is a filling factor. The circuit is stable within 1 Hz, the enhanced nuclear susceptibility producing changes of up to 14 kHz.

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3. EXPERIMENTAL RESULTS AND DISCUSSION.- First, the temperature dependence of the susceptibility was measured between 1.2 K and 15 mK. Since the electronic susceptibility of HoVO$_4$ is temperature independent in the helium range, the observed linear dependence of frequency upon inverse temperature down to 25 mK was associated with the enhanced nuclear susceptibility. The susceptibility was used as a thermometer for the crystal in the range 20<T<40 mK.

Starting from an initial temperature of 35 mK, demagnetisations were made from various initial fields applied parallel to the crystal a axis, to zero final field. The measured residual field of the solenoid was less than 20 gauss. For runs with small initial fields, the observed susceptibility was used to give the final temperature. Using the relation $T_f = (B^2 + b^2)^{1/2}$, we obtain the effective residual field $B_0 = 466$ gauss. Contributions to $B_0$ arise from the quadrupole interaction, equivalent for $^{165}$Ho to a field $B_Q = 2P/\gamma_L = 339$ gauss, and from the spin-spin interaction field b. Taking $B^2 = B_Q + b^2$ we find $b = 320$ gauss. With these parameters theoretical calculations of the entropy and final temperature of the enhanced nuclear system were made, for demagnetisations from higher initial fields. The measured susceptibility following demagnetisation is shown in figure 1, plotted against calculated final temperature.

In zero field the nuclear orientation results show that the nuclear spins are equally divided between antiferromagnetic domains directed along the a and a' axes, and a perpendicular a' axis. Thus we expect the zero field susceptibility along the z axis below $T_N$ to be of the form $\chi_{//}$ and $\chi_{\perp}$, as given in figure 1. The agreement with experiment is good, lending support to both the model and the temperature calibration. On increasing the applied field to 60 gauss, the a' axis domains, being the spin-flop configuration, predominate. At about 140 gauss the susceptibility is larger than the maximum observed in zero field. This indicates a rapidly changing magnetisation which we interpret as spin flip transition to the ferromagnetic phase.

At 400 gauss the tail of the spin-flip transition is seen at the lowest temperatures reached. For higher fields all spins are polarised along the field direction resulting in the observed reduction of susceptibility.
These spin structures also explain the main features of the nuclear orientation results which will be discussed more fully elsewhere.

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References