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MAGNETIC EXCITATIONS IN THULIUM

K. A. McEwen (1) and U. Steigenberger (2)

(1) Department of Physics, Birkbeck College, University of London, Malet Street, London WC1E 7HX, G.B.
(2) Neutron Division, Rutherford Appleton Laboratory Chilton, Didcot, Oxon OX11 0QX, G.B.

Abstract. - We have observed, for the first time, magnetic excitations in a single crystal of thulium, using inelastic neutron spectroscopy. In the ferrimagnetic phase three modes with energies around 4 meV, 8 meV and 15 meV were clearly seen. We have considered our results in terms of crystal field level schemes for thulium.

The rare earth metal thulium (hcp, electronic configuration 4f\( ^{12} \)) exhibits a particularly interesting sequence of magnetic phases. Below \( T_N = 56 \) K, the moments are sinusoidally modulated along the hexagonal axis [1, 2]. This structure “squares up” at lower temperatures (below 40 K) as the amplitude begins to exceed the free-ion moment of \( 7 \mu_B \). Measurements of the temperature dependence of the magnetic periodicity show that there is a first-order lock-in transition at 32 K to an exact 7-layer periodicity. Below this temperature the structure is ferrimagnetic and comprises 4 layers of moments parallel to the c-axis, followed by 3 antiparallel layers. The ferrimagnetic coupling persists to the lowest temperatures.

We performed a first study of the magnetic excitations in a single crystal of thulium using the triple-axis spectrometer TAS 3 at Riso National Laboratory. The single crystal was prepared by J. S. Abell at the Department of Metallurgy, University of Birmingham. Measurements were made at 12 K (in the ferrimagnetic phase) and at higher temperatures. The instrument was operated in the constant-\( k_F \) mode (\( k_F = 2.662 \) Å\(^{-1} \)). Figure 1 shows an energy scan for \( Q = (1, 0, 1) \) as a function of temperature. There is clear evidence for a mode at about 3.5 meV. The energy of the peak changes little with increasing temperature, whereas the intensity steadily decreases. At 80 K, above the \( N\)éel temperature, the magnetic scattering has all been shifted towards the elastic line and manifests itself as a broad tail indistinguishable from the elastic scattering.

Further studies were made at the Institute Laue-Langevin using the IN8 triple-axis spectrometer. The experimental configuration (\( k_F = \) constant =2.662 Å\(^{-1} \), collimation: 40° – 60° – 60°) was comparable to that at Riso. We performed constant-Q scans along the [001] and [101] directions. For some scans at high energy transfer we also used the Cu111 monochromator with \( k_F = 4.189 \) Å\(^{-1} \). Figure 2 shows a scan at 5 K for \( Q = (1, 0, 2.5) \) with energy transfers up to 20 meV. Three modes at 4.3 meV, 8 meV and 15.3 meV were clearly seen. We have considered our results in terms of crystal field level schemes for thulium.
clearly observed. In figure 3 the \( Q \) dependence of the lower energy modes is shown along the [0, 0, 1] direction. For some \( Q \) values there seems to be evidence for a further mode located at about 5-6 meV. Because of the magnetic orientation factor in the neutron scattering cross-section, the modes observed along the \( c \)-axis must have a predominantly transverse character. The excitations are considerably broader than the instrumental resolution (1 meV). We attribute this to the effect of the site-dependent molecular field in the ferrimagnetic phase.

In our measurements we found generally only little dispersion, which implies a rather weak exchange coupling. We have therefore attempted to describe our results in terms of a crystal field model instead of the standard spinwave model used for the other heavy rare earth. As a starting point we used the crystal field parameters derived by Touborg [3] from susceptibility measurements on dilute Tm alloys and employed by Fynbo [4] to describe his magnetisation measurements. This model predicts a strong transverse transition at 5.5 meV and two weak excitations at 11.3 meV (transverse) and at 12.3 meV (longitudinal). With increasing molecular field the ground state becomes an almost pure \([-6] \) state and the matrix elements for the two higher transitions become very small. The predictions of this model are obviously in contradiction with our experimental results. We attempted to obtain a better fit constraining only the second order parameter \( D_0 \). This parameter can be deduced from the uniaxial anisotropy of the susceptibility [5]. The value calculated in this way is \(-0.075 \) meV, about 15 \% less than that used by Fynbo [4]. We find that there is a certain range for the remaining parameters \( B_4, B_6 \) and \( B_8 \) where the ground state and first excited states are the symmetric \([\pm 3] \) and the \([-6] \) states, separated only by a small energy gap. This means that both levels could be thermally occupied at 5 K and consequently transitions from both states into appropriate higher energy states are possible. However, no parameter set could be found that describes in a satisfactory way both the energies of the transitions and their relative intensities. We also allowed for a variation in the local mean field, since different layers of spins experience magnetically different surroundings. We find it puzzling that our experimental results cannot be readily explained within a simple crystal field model.

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