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Spin waves in the ferrimagnetic and ferromagnetic phases of thulium

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The magnetic excitations in both the ferri- and ferromagnetic phases of thulium have been measured. The dispersion in the ferromagnetic phase is much larger than predicted from the exchange interaction derived from the ferrimagnetic phase. Magnetoelastic interactions have a significant effect on the excitations in both phases, and on the ferri–ferromagnetic transition field.

1. Introduction

The rare earth metal thulium exhibits a particularly interesting sequence of magnetic phases. Below $T_N = 57$ K, the moments are sinusoidally modulated along the hexagonal c-axis. This structure "squares up" below 40 K. At 32 K there is a first-order lock-in transition to an exact 7-layer periodicity. Below 32 K, the structure is ferrimagnetic and comprises 4 layers of moments parallel to the c-axis, followed by 3 antiparallel layers. Thulium becomes ferromagnetic when the internal magnetic field along the c-axis exceeds 2.8 T.

We have measured the excitations in both the ferri- and the ferromagnetic phases of thulium in order to compare the exchange interaction $\mathcal{J}(q)$. Since Tm has $J = 6$ and $g = 7/6$, the net magnetic moment per atom increases from $1\mu_B$ to $7\mu_B$ between the two phases. This modifies significantly the splitting of the spin-up and spin-down conduction electron bands and we observe dramatic changes in the excitation spectra. A detailed experimental study and analysis of the spin waves along the c-direction in the ferrimagnetic phase has recently been reported by us [1]. In this paper we concentrate on measurements in the basal plane.

2. Experimental details

A large single crystal (1.5 cm^3) of very high quality was grown for us by the Ames Laboratory. The experiments were performed at the Institut Laue–Langevin using the IN20 triple-axis spectrometer with constant $k_f = 2.662$ Å and collimations 40′-60′-60′. The instrumental resolution, measured with vanadium, was 1.0 meV. The crystal was mounted with its c-axis vertical, enabling us to measure the excitations propagating in the basal plane. A magnetic field could be applied parallel to the c-direction.

3. Experimental results

We measured carefully the intensity of the ferromagnetic Bragg peak at $\boldsymbol{Q} = (1, 0, 0)$ to monitor the phase transition. At $B = 3.2$ T, the overall peak intensity, which contains nuclear and ferromagnetic contributions, increased by a factor of 2.4 and at $B = 4.5$ T by 2.8. At this field the intensity had apparently saturated. The very large demagnetisation field in thulium means that the transition into the ferromagnetic phase is only accomplished after an increase of about 1 T in the applied field. This effect has already been observed in magnetostriction measurements [2] which demonstrated that an applied field of 3.6 T was necessary to complete the transition.

Figure 1 shows the development of the excitation spectrum at $\boldsymbol{Q} = (2, -1, 0)$ at a temperature of 7 K. At zero field we observe a pronounced inelastic peak centred at 8.5 meV which is somewhat broader than the instrumental resolution. A slight decrease in intensity was observed at 2.4 T, i.e. still below the critical magnetic field of $B = 2.8$ T. When the field is increased to 3.2 T the initially broad peak splits into two with roughly equal intensity, indicating that the system is in a mixed state. On increasing the magnetic field to 4.5 T the peak positions remain unchanged but the zero-field peak has almost disappeared. We attribute the weak higher energy peak to the presence of residual ferrimagnetic domains, since the structure factor of the Γ-point is entirely acoustic in the ferromagnetic phase. Several other high symmetry points and directions were investigated and these results will be published elsewhere in greater detail [3].

In fig. 2 we present the dispersion curve for thulium along the $\boldsymbol{a}^*$ direction, i.e. from M to Γ, in the ferromagnetic phase at a temperature of 7 K ($B = 4.5$ T). The two branches are clearly separated by an energy difference of about 2.5 meV at the zone centre.

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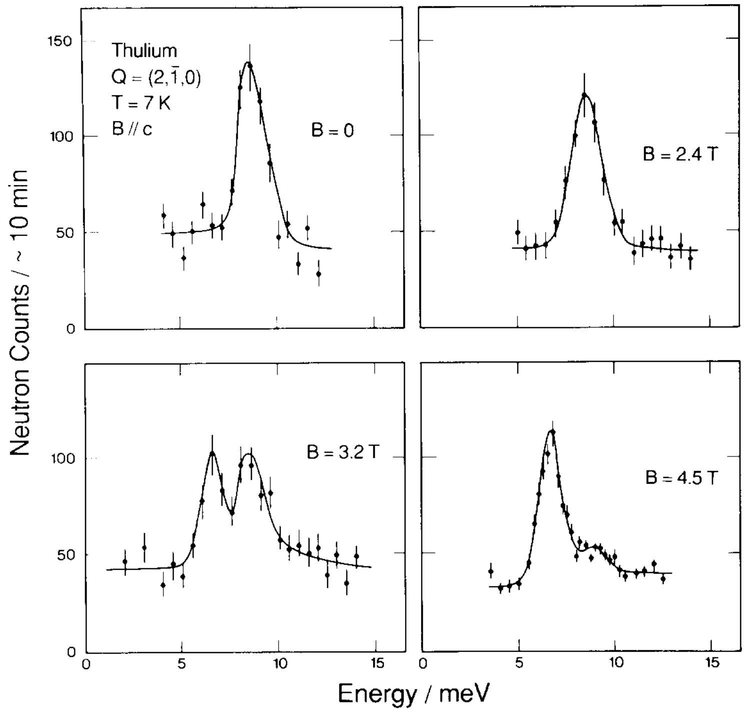

Fig. 1. The excitation spectrum at $Q = (2, -1, 0)$ for various magnetic fields along the c-axis.

At $Q = (0.8, 0, 0)$ there appears to be crossing of the two branches and the energy difference at the zone boundary has about the same value as at the zone centre.

4. Discussion

According to the model developed by Jensen in ref. [1], the crystal field splitting $\Delta_{CF} = 8.5$ meV and the dispersion $J[\mathcal{J}(0) - \mathcal{J}(2\pi/c)] = 0.7$ meV. The energy

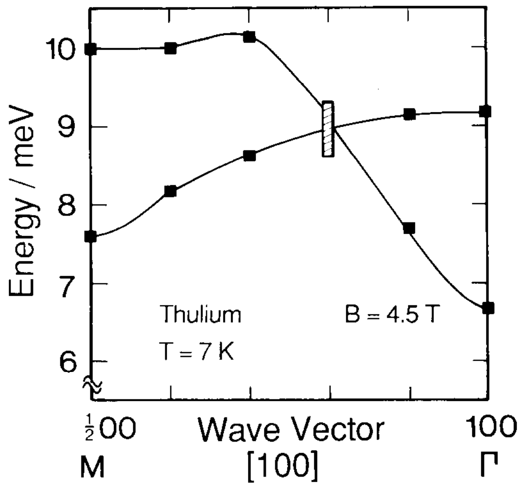

Fig. 2. The dispersion relation of the magnetic excitations from M to Γ in the ferromagnetic phase of thulium.

of the acoustic spin wave in the ferromagnetic phase at the Γ point should then be $E_A = \Delta_{CF} + g\mu_B H = 8.8$ meV in a field of 4.5 T, and the optical mode energy should be $E_O = (8.8 + 0.7)$ meV $= 9.5$ meV, whereas the observed values are $E_A = 6.7$ meV and $E_O = 9.2$ meV.

Results from the magnetostriction measurements [2] indicate a surprisingly large expansion (0.7%) of the c-axis at the phase transition. This immediately explains the large discrepancy in ref. [1] between the calculated transition field of 4.2 T and the experimental value of 2.8 T, as the magnetoelastic energy related to this change of the c-axis (neglecting a possible modification of the basal plane) corresponds to a reduction of the calculated transition field by 1.3 T.

Our results for E_A and E_O show that the two-ion interactions change considerably between the two phases. The magnitude of the energy changes are comparable with that due to the c-axis expansion. They may be explained in a phenomenological way by a two-ion magnetoelastic coupling like the one considered in Er in connection with its transition to the cone phase (see the discussion in section 2.3 of [4]). This magnetoelastic coupling is not directly related to the one which mixes the spin waves and the transverse phonons in Tm at zero field [1]. The magnon-phonon coupling may be explained as a crystal-field effect with a magnitude which compares reasonably well with that observed in other heavy rare earths. However, the present magnetoelastic coupling must be of two-ion origin, and it has to be somewhat stronger than the corresponding coupling derived in Er. Moreover, it has to be extremely anisotropic in order to explain the reduction of the effective value of Δ_{CF} by about 2.1 meV. It is possible that the drastic changes of the Fermi surface of the conduction electrons occurring at the ferromagnetic transition may affect all the magnetic couplings in Tm in a more direct way than that described by a magnetoelastic coupling. Further details will be published in due course.

References

[1] K.A. McEwen, U. Steigenberger and J. Jensen, Phys. Rev. B 43 (1991) 3298.

[2] S. Zochowski and K.A. McEwen, J. Magn. Magn. Mater., to be published.

[3] K.A. McEwen, U. Steigenberger, J. Jensen and J.L. Martinez, to be published.

[4] J. Jensen and A.R. Mackintosh, Rare Earth Magnetism: Structures and Excitations (Oxford University Press, Oxford, 1991).