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MAGNETIC EXCITATIONS IN THULIUM METAL

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ABSTRACT

We have performed inelastic neutron scattering measurements on a single crystal specimen of Tm at wavevectors $\vec{\kappa} = (1, 1, \zeta)$ and $(0, 0, 2 + \zeta)$ ($\zeta = 0, ..., 1$). Most of the measurements have been made at $T = 5\,K$, where Tm exhibits a seven layer ferrimagnetic-antiphase-domain structure (four moments up, parallel to the c-axis, followed by three moments down). At this temperature the excitation spectra consist of three peaks. The two lower energy excitations have been identified as originating from magneto-vibrational scattering from the TA phonon, while the higher energy excitation is magnetic and exhibits only a weak dispersion (between 8.3 and 9.6 meV). At $T = 50\,K$, a temperature at which the system exhibits a c-axis sinusoidally modulated structure, the magnetic mode shows significant softening and broadening. The magneto-vibrational scattering vanishes above the Nèel temperature ($T_N = 58.5\,K$) while the magnetic mode persists at least up to $T = 70\,K$. These results suggest that the Hamiltonian in this system is dominated by the crystal-field-anisotropy energy, and that the exchange interaction is relatively weak.

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1. INTRODUCTION

In the last 25 years very detailed studies of the magnetic excitations of most of the heavy rare earth elements have been published.[1] These studies, which were performed utilizing neutron scattering techniques, have greatly contributed to our present understanding of the interactions that determine the complicated magnetic structures found in these systems. The study of the magnetic excitations in Tm, on the other hand, has been hampered by the lack of large high-quality crystals of this element and it is only in very recent years that efforts to perform this study have been undertaken.[2-3] In this paper we report our progress in this study.

Thulium is the last of the magnetic heavy rare elements. It has an *hcp* structure and exhibits a very unusual series of magnetic structures.[4] Below the Nèel temperature T_N the magnetic moments order in a c-axis sinusoidally modulated phase. Upon cooling the sinusoidal structure gradually squares up and below a temperature T_c it develops a ferrimagnetic component and the system locks in a seven-layer ferrimagnetic antiphase-domain structure (four moments up, parallel to the c-axis, followed by three moments down).

The nature of the magnetic excitations in the heavy rare earth elements is determined by the Hamiltonian [1]:

$$\mathcal{H} = \mathcal{H}_{ex} + \mathcal{H}_{cf}. \tag{1}$$

In this equation $\mathcal{H}_{ex}$ is the exchange energy, which is due to a long-range oscillatory exchange interaction between highly localized $4f$ moments. It is predominantely isotropic and has a Heisenberg form. The second term, $\mathcal{H}_{cf}$, is the crystal-field anisotropy energy. Other sources of anisotropy (magnetoelastic energy, etc) are neglected in this Hamiltonian. It is frequently assumed that in the heavy rare earth elements $\mathcal{H}_{cf} << \mathcal{H}_{ex}$ and that the only effect of the crystal-field anisotropy is to impose a preferred direction of the magnetization. The energy levels under this assumption are equally-spaced J_z levels and the elementary excitations of the system are spin waves, or collective propagating excitations from the $< J_z >= J$ ground state. In this model the dispersion relation can be obtained by means of Holstein-Primakoff-type transformations followed by the proper diagonalization of the transformed Hamiltonian. This approach has been used succesfully to describe the magnetic excitations of Gd, Tb, Dy and Ho.[1]

When $\mathcal{H}_{cf} \approx \mathcal{H}_{ex}$, on the other hand, the energy diagram of the system does not consist of equally-spaced J_z levels and Holstein-Primakoff-type transformations are not applicable. In this case a transformation that properly includes the effect of the crystal field on the energy diagram must be carried out. This has been performed by Lingård and Lingård and Cooke [5] to obtain the dispersion relation of Er. In the case where $\mathcal{H}_{cf} > \mathcal{H}_{ex}$ the energy diagram consists essentially of crystal-field levels and the exchange imposes a small dispersion on the excitations. In the extreme case where $\mathcal{H}_{ex} = 0$ the energy diagram consists only of crystal-field levels. The study of the magnetic excitations of Tm is of interest because this is a system in which the crystal-field anisotropy is expected to be at least comparable to the exchange interaction.

2. EXPERIMENTAL

The sample used in our experiments was a high-quality single crystal of Tm prepared at Ames Laboratories, Ames, Iowa. The neutron scattering measurements were performed with a conventional triple axis spectrometer at the National Institute of Standards and Technology (formerly National Bureau of Standards) reactor. The measurements were performed in the constant scattering wavevector $\vec{\kappa}$ mode at $\vec{\kappa} = (1, 1, \zeta)$ and $(0, 0, 2 + \zeta)$ (for $\zeta = 0, ..., 1$) with a fixed final neutron energy $E_f = 14.8$ meV. A pyrolitic graphite filter was used in order to eliminate higher-order wavelength contaminations. Soller-slit collimators were utilized to obtain horizontal collimations of 40'-40'-40'-40'.

The c-axis sinusoidally modulated structure of Tm is characterized by a wavevector $\vec{Q} \approx (0, 0, 2/7)$. The Nèel temperature of the specimen under study, measured as the temperature at which the first magnetic satellite at $\vec{\kappa} = (1, 1, 0) \pm \vec{Q}$ appears, was found to be $T_N = 58.5\,K$. Most of the measurements were performed at $T = 5\,K$, a temperature at which Tm orders in an antiphase-domain ferrimagnetic structure of characteristic wavevector $\vec{Q} = (0, 0, 2/7)$. At this temperature magnetic satellites are observed at $\vec{\kappa} = \vec{\tau} \pm \vec{Q}$ (where $\vec{\tau}$ is a reciprocal lattice vector) as well as at the higher harmonics $\vec{\kappa} = \vec{\tau} \pm (0, 0, 4/7)$ and $\vec{\kappa} = \vec{\tau} \pm (0, 0, 6/7)$.[4]

The neutron scattering processes that are of interest in our study are: magnon creation, phonon creation (through neutron-nuclear interaction) and magneto-vibrational scattering (phonon creation through neutron-magnetic interaction). The neutron cross section for the creation of a magnon of wavevector $\vec{q}$ and energy E_q in a periodic magnetic structure of characteristic wavevector $\vec{Q}$ is[1,6]:

$$\frac{d^2\sigma}{d\Omega\, dE_M} = \gamma(\vec{\kappa})\frac{k_f}{k_i}\frac{1}{2J}[n(E_q)+1]\sum_{\vec{\tau}}\{\frac{1}{4}(1+e_Q^2)\,A(\vec{q})\,\delta(\vec{\kappa}-\vec{q}\pm\vec{Q}-\vec{\tau})$$

$$+(1-e_Q^2)\,B(\vec{q})\,\delta(\vec{\kappa}-\vec{q}-\vec{\tau})\}\,\delta(E-E_q). \qquad (2)$$

In this equation $\gamma(\vec{\kappa})$ is a coefficient that contains the magnetic and geometrical form factors, and $A(\vec{q})$ and $B(\vec{q})$ are coefficients that depend on Fourier transforms of the exchange. The other symbols are those of standard usage: k_f and k_i are the final and initial wavevectors of the neutrons, $n(E_q)$ is the Bose thermal population factor, $\vec{e}$ is a unit vector in the direction of $\vec{\kappa}$ and e_Q its component along $\vec{Q}$. Note from this cross section that an excitation of energy E_q should be observed at $\vec{\kappa} = (\vec{q} + \vec{\tau})$ as well as at $\vec{\kappa} = (\vec{q} + \vec{\tau} \pm \vec{Q})$ indicating that the magnon wavevector $\vec{q}$ is measured from a reciprocal lattice point $\vec{\tau}$ or from the magnetic satellites. The neutron cross section for magnetovibrational scattering in a periodic structure of characteristic wavevector $\vec{Q}$ is[6]:

$$\frac{d^2\sigma}{d\Omega\, dE_{MV}} = \gamma(\vec{\kappa})\frac{k_f}{k_i}\left(\frac{J(T)}{J}\right)^2[n(E_q)+1]\frac{\hbar^2\kappa^2(\vec{e}\cdot\vec{\xi}_q)^2}{2M\,E_q}$$

$$\times\sum_{\vec{\tau}}\frac{1}{2}\,\delta(\vec{\kappa}-\vec{q}\pm\vec{Q}-\vec{\tau})\,\delta(E-E_q). \qquad (3)$$

Because this process is just the creation of a phonon through the neutron-magnetic interaction, its cross section is very similar to that for a phonon creation (through neutron-nuclear

interaction): it is proportionl to $\kappa^2(\vec{e}.\vec{\xi}_q)^2 \,/\, E_q$, where $\vec{\xi}$ is the polarization vector of the phonon. Note, however, that unlike for the usual phonon case this cross section depends on the magnetic form factor, which is included in $\gamma(\vec{\kappa})$, and on the magnetization. Also unlike the phonon case the wavevector $\vec{q}$ is measured from the magnetic satellites.

The above cross sections indicate that in a neutron scattering experiment we should expect to observe a variety of branches in the dispersion relation: phonon and magnon branches from the reciprocal lattice points, and magnon and magnetovibrational scattering branches from each magnetic satellite.

3. RESULTS AND DISCUSSION

The low-temperature ($T = 5K$) excitation spectra for Tm at all wavevectors under study consisted of three peaks. This can be seen in figure 1, which shows constant-$\vec{\kappa}$ scans at $\vec{\kappa} = (1, 1, \zeta)$ for $\zeta = 0,\ 0.3,\ 0.8$. The solid lines in this figure are the result of least-squares fits to three Gaussians. The excitation spectra along the $(0, 0, 2 + \zeta)$ direction were qualitatively similar to those of figure 1 but the two lower-energy excitations were weaker. These spectra are also in qualitative agreement with those reported by McEwen and Steigenberger[2] in the $0 - 12$ meV range. We found, however, no evidence of the excitation at 15 meV reported by these authors.

In all cases the two lower-energy excitations are sharp and show a significant dispersion. The higher-energy excitation, on the other hand, exhibits only a weak dispersion (between 8.3 and 9.6 meV) and a linewidth that is wavevector dependent. In figure 2 all the excitation energies identified from scans along the $(1, 1, \zeta)$ and $(0, 0, 2 + \zeta)$ directions have been plotted *vs.* ζ. In this figure we have also plotted (in solid lines) the expected dispersion relation for the TA phonon (branch from $\zeta = 0$) as well as the dispersion relations for magnetovibrational scattering from the TA phonon (branches from the magnetic satellites at $\zeta = \pm 2/7, -4/7$, the branches for $\zeta = 4/7, \pm 6/7$ have been omitted). Because no phonon dispersion data are available for Tm the dispersion relation shown in this figure is that for Tb (of comparable mass and same structure) reported by Houmann and Nicklow.[7] The dispersion relations for the magnetovibrational scattering shown in the same figure have been obtained by translating the phonon dispersion relation from $\zeta = 0$ to $\zeta = \pm 2/7, -4/7$. The striking agreement of the lower-energy excitations and the solid lines strongly suggests that these should be identified as a TA phonon and magnetovibrational scattering from the TA phonon. We must remark that in principle the TA phonon should not be observed along the $(0, 0, 2 + \zeta)$ direction due to the $(\vec{e}.\vec{\xi}_q)^2$ dependence of the cross section of equation (3). In practice, however, it is not unusual that transverse phonons are observed in purely longitudinal scans due to multiple scattering processes (*i.e.* the creation of a phonon followed or preceded by Bragg scattering). The intensities of these lower-energy excitations along the $(1, 1, \zeta)$ direction are consistent with the $\kappa^2(\vec{e}.\vec{\xi}_q)^2 \,/\, E_q$ dependence of the cross section of equation (3). Also the temperature dependence of these excitations, observed by McEwen and Steigenberger[2] and ourselves, seems to be consistent with this cross section: the excitations that we identify as magnetovibrational scattering depend on the magnetization, which becomes smaller at higher temperatures and vanishes at the Néel temperature.

These observations suggest that only the higher-energy branch shown in figure 2 corresponds to magnetic excitations. A recent neutron scattering experiment performed with

polarization analysis has confirmed this interpretation.[8]

The limited dispersion of this branch suggests that the crystal-field anisotropy term of the Hamiltonian is dominant over the exchange, and that the observed gap in the dispersion relation (≈ 8 meV) may be closely related to the first dipolar transition in the crystal-field-only levels scheme. The energy of this transition can be estimated using the crystal-field parameters estimated by Touborg[9] from paramagnetic susceptibility measurements on dilute Tm alloys and pure Tm. This value is 5.5 meV (from the dilute alloys estimations) or 7.7 meV (from the pure Tm estimations).

Because these excitations are "magnons" (or magnon-like) the cross section of equation (2) suggests that several branches of the dispersion relation should be observed (from the reciprocal lattice point and from each satellite, like in the case of the magnetovibrational scattering discussed above). If such branches exist we have not been able to resolve them due to our relatively coarse energy resolution ($\Delta E_{FWHM} = 1.4$ meV at 8 meV). The fact that these excitations are broader than the energy resolution and that their linewidths are wavevector dependent, on the other hand, suggests that such an interpretation of the data is not unreasonable. Upon warming these magnetic modes soften and become broader. These broad modes persist at least up to $T = 70\,K$. Further measurements are in progress.

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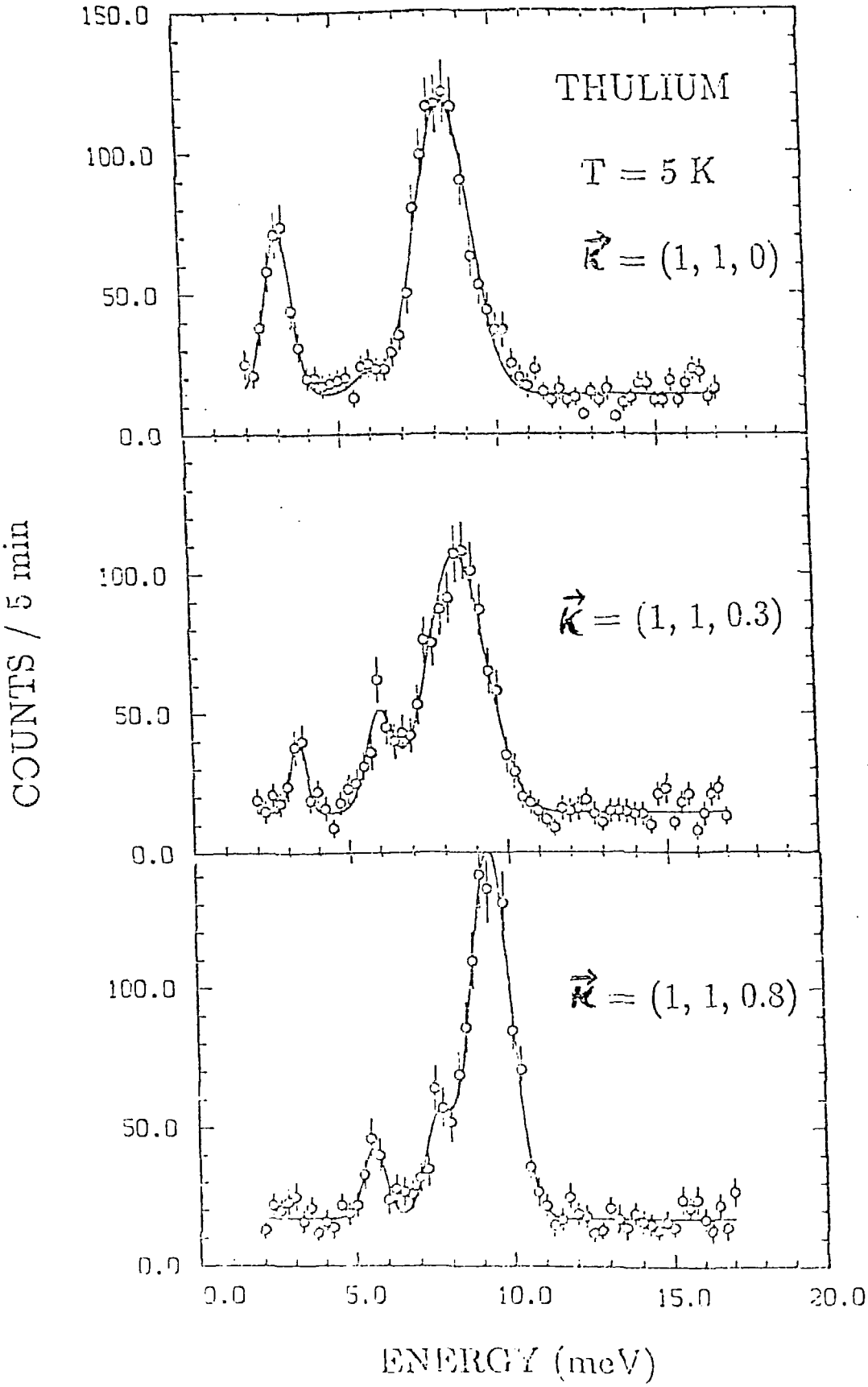
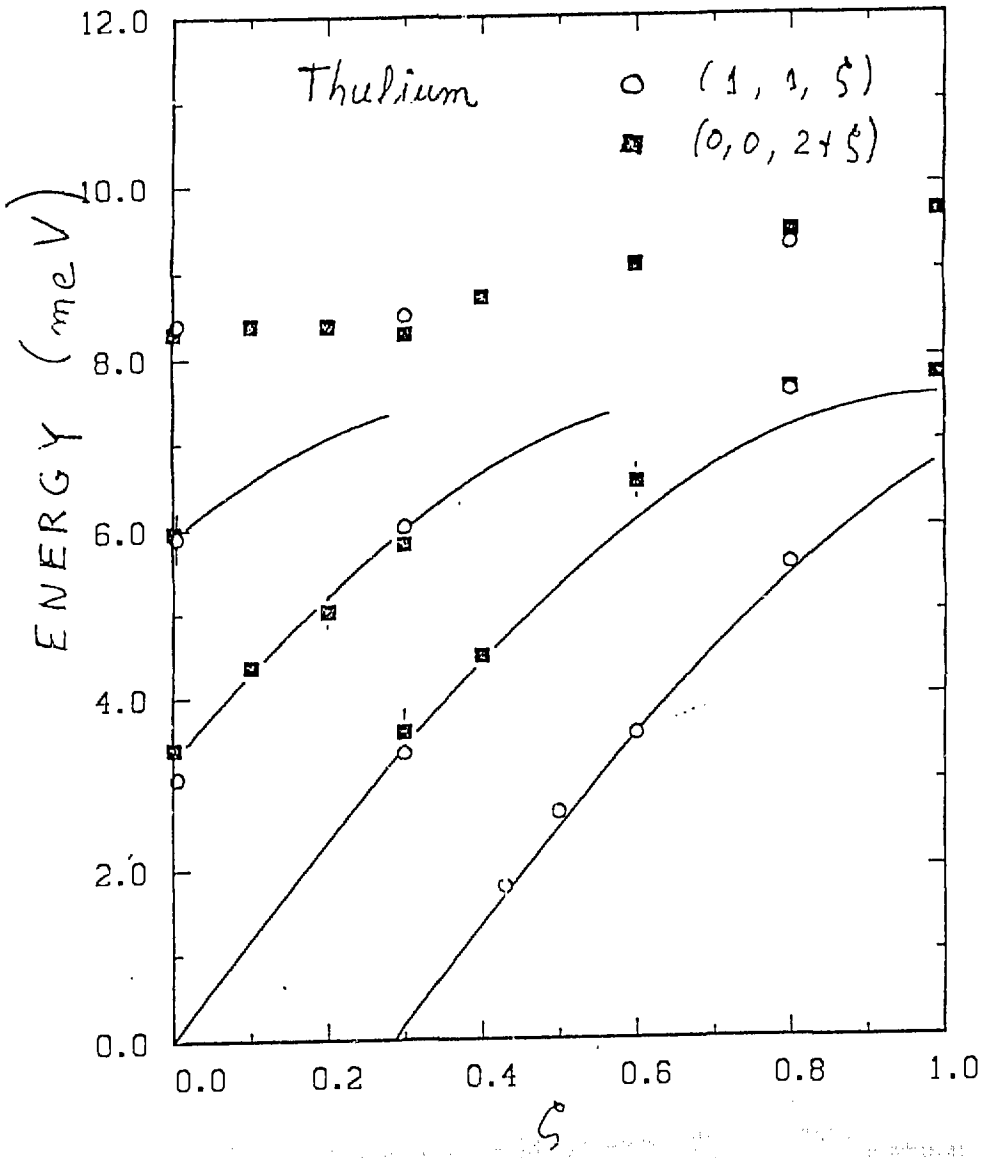
FIGURE CAPTIONS

FIGURE 1. Constant-$\vec{\kappa}$ scans at $\vec{\kappa} = (1, 1, \zeta)$ ($\zeta = 0, 0.3, 0.8$) for Tm at $T = 5\,K$. In all cases three excitations were observed, the lower energy ones show significant dispersion while the higher energy one exhibits only a weak dispersion. The solid lines are from a fit to three gaussians.

FIGURE 2. Dispersion relations of the observed excitations in Tm along the $(1, 1, \zeta)$ (open circles) and $(0, 0, 2 + \zeta)$ (full squares) directions. The solid lines correspond to the dispersion relation expected for the creation of a TA phonon (through nuclear interaction) as well as for magneto-vibrational scattering from the TA phonon.

FIGURE 1


150.0
100.0
50.0
0.0
THULIUM
T = 5 K
$\vec{K}$ = (1, 1, 0)
100.0
50.0
0.0
$\vec{K}$ = (1, 1, 0.3)
100.0
50.0
0.0
$\vec{K}$ = (1, 1, 0.8)
0.0
5.0
10.0
15.0
20.0
COUNTS / 5 min
ENERGY (meV)


Thulium
(1, 1, ζ)
(0, 0, 2+ζ)
ENERGY (meV)
12.0
10.0
8.0
6.0
4.0
2.0
0.0
0.0
0.2
0.4
0.6
0.8
1.0
ζ