Neutron-scattering study of the magnetic excitations of thulium metal

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We have performed neutron-scattering experiments to study the magnetic excitations of Tm in its low-temperature ferrimagnetic-antiphase-domain (FAD) phase. Our measurements confirm our earlier finding that the excitation spectrum of Tm in its FAD phase consists of one magnon mode with limited dispersion (8.3–9.6 meV). This result suggests that the Hamiltonian is dominated by the crystal-field-anisotropy energy and that the exchange interaction is relatively weak. Two additional lower-energy excitations have also been observed, and they have been identified as originating from magnetovibrational scattering from the TA phonon.

The study of the magnetic excitations of the heavy rare earths has been a topic of considerable interest during the last 25 years.[1] It is only very recently, however, that the availability of large high-quality single crystals of Tm has made possible the study of the magnetic excitations of this element using neutron-scattering techniques. There have been at least two independent preliminary studies of this type,[2,3] which have resulted in conflicting results. McEwen and Steigenberger (MS)[2] have reported that the magnetic excitation spectrum of Tm at $T=4.5$ K, a temperature at which this system orders in a seven-layer ferrimagnetic-antiphase-domain (FAD) structure, consists of three modes at around 4, 8, and 15 meV. Based on the fact that these modes exhibited very little dispersion, these authors attempted, unsuccessfully, to describe them in terms of a crystal-field model. In contrast, our previous measurements on Tm at $T=5$ K showed that there is only one magnetic mode at around 8 meV with a weak dispersion.[3] In order to resolve the disagreement concerning the nature of the elementary magnetic excitations of Tm in the FAD phase, we have performed neutron-scattering measurements using both unpolarized and polarized neutrons. In this paper we report the results of these new measurements.

Thulium metal has an hcp structure and as in the rest of the heavy rare-earth metals, its magnetic moment is due to an incompletely filled $4f$ electronic shell which is well shielded by outer closed $5s$ and $5p$ shells. These $4f$ electrons form a ground state of total angular momentum $\mathbf{J}$ ($J=6$) with a saturation moment per ion $gJ=7\mu_B$. As a result of the competition between the indirect (RKKY) exchange interaction (which favors periodic structures) and the crystal-field anisotropy (which tends to align the moments in unique crystallographic directions), Tm exhibits a very unusual series of magnetic structures.[4] Below $T_N=58.5$ K, its magnetic moments order in a c axis sinusoidally modulated phase, characterized by a wave vector $\mathbf{Q}\approx(0,0,2/7)$. Upon cooling, this sinusoidal structure gradually squares up and, below $T_c=32$ K, it develops a ferrimagnetic component and the system locks in a seven-layer FAD structure (four moments up, parallel to the c axis, followed by three moments down), characterized by a wave vector $\mathbf{Q}_{\rm FAD}=(0,0,2/7)$.[4,5]

The sample used in our experiments was a high-quality single crystal of Tm prepared at the Ames Laboratory. The unpolarized neutron-scattering measurements were performed on a conventional triple-axis spectrometer at the reactor NBSR at the National Institute of Standards and Technology. For these measurements pyrolytic graphite (PG 002) crystals were used as monochromator and analyzer with Soller-slit collimations of 40′-40′-40′-40′. A PG filter was placed after the sample in order to eliminate higher-order wavelength contaminations. The polarized neutron measurements were performed on a triple-axis spectrometer with full polarization analysis at the NRU reactor at Chalk River Nuclear Laboratories. For these measurements Heusler alloy polarizing crystals were used as monochromator and analyzer, and a PG filter and spin-flipper were positioned between the sample and the analyzer. The effective beam divergence determined by distance collimation was 40′ before the sample and 1.8° after the sample. In order to obtain a single magnetic domain for the polarization analysis measurements, the sample was cooled in a horizontal magnetic field of 1 T applied along its c axis with a special magnet cryostat[6] which allows almost 360° access for neutron beams. When the temperature reached 4 K, this field was reduced to 0.5 T and maintained at this value throughout the experiment. The constant scattering-wave-vector measurements with unpolarized neutrons were performed at $\boldsymbol{\kappa}=(1,1,\zeta)$ and $(0,0,2+\zeta)$ (for $\zeta=0,\ldots,1$) with a fixed final neutron energy $E_f=14.8$ meV, and those with polarized neutrons at $\kappa=(1,1,0)$ with $E_f=14.6$ meV.

The inelastic neutron-scattering processes that are of

interest in our study are magnon creation, phonon creation (through neutron-nuclear interaction), and magnetovibrational scattering[7] (phonon creation through neutron-magnetic interaction). The unpolarized-neutron cross section for the creation of a transverse magnon of wave vector $\mathbf{q}$ and energy E_q in the FAD phase of Tm is

$$\frac{d^2\sigma}{d\Omega\, dE_f}=\gamma(\boldsymbol{\kappa})A(\mathbf{q})\frac{k_f}{k_i}(1+|\hat{\kappa}_Q|^2)[n(E_q)+1]\times\sum_{N=0}^{3}\sum_{\tau}\delta(\boldsymbol{\kappa}-\mathbf{q}\pm N\mathbf{Q}_{\mathrm{FAD}}-\boldsymbol{\tau})\delta(E-E_q)\ . \tag{1}$$

In this equation $\gamma(\boldsymbol{\kappa})$ is a coefficient that contains the magnetic form factor, $|\hat{\kappa}_Q|$ is the component of the unit vector $\hat{\kappa}$ along $\mathbf{Q}_{\mathrm{FAD}}$, and $A(\mathbf{q})$ gives the strength of the magnon. The other symbols are those of standard usage: k_f and k_i are the final and initial neutron wave vectors, $n(E_q)$ is the Bose thermal population factor, and $\boldsymbol{\tau}$ is a reciprocal lattice vector. For Tm in the FAD phase, magnetic satellites can be observed at wave vectors $\boldsymbol{\kappa}=\boldsymbol{\tau}\pm N\mathbf{Q}_{\mathrm{FAD}}$ for $N=1,2,3$.[4,5,8] Note from Eq. (1) that a magnon of energy E_q should be observed at $\boldsymbol{\kappa}=(\mathbf{q}+\boldsymbol{\tau}\pm N\mathbf{Q}_{\mathrm{FAD}})$ for $N=0,\ldots,3$. Thus the magnon wave vector $\mathbf{q}$ is measured either from a reciprocal lattice point $\boldsymbol{\tau}$ (when $N=0$) or from a magnetic satellite (when $N=1,2,3$). The unpolarized-neutron cross section for magnetovibrational scattering in the FAD phase of Tm is

$$\frac{d^2\sigma}{d\Omega\, dE_f}=\gamma(\boldsymbol{\kappa})\frac{k_f}{k_i}(1-|\hat{\kappa}_Q|^2)\sum_{N=0}^{3}\sum_{\tau}\langle J^z(N\mathbf{Q}_{\mathrm{FAD}})\rangle_T^2[n(\mathscr{E}_q)+1]\frac{\hbar^2(\boldsymbol{\kappa}\cdot\boldsymbol{\xi}_q)^2}{2M\mathscr{E}_q}\delta(\boldsymbol{\kappa}-\mathbf{q}\pm N\mathbf{Q}_{\mathrm{FAD}}-\boldsymbol{\tau})\delta(E-\mathscr{E}_q)\ . \tag{2}$$

Because this process is just the creation of a phonon of energy $\mathscr{E}_q$ through the neutron-magnetic interaction, its cross section is very similar to that for a phonon creation through neutron-nuclear interaction. Thus the scattering is proportional to $(\boldsymbol{\kappa}\cdot\boldsymbol{\xi}_q)^2/\mathscr{E}_q$, where $\boldsymbol{\xi}_q$ is the polarization vector of the phonon. Note, however, that unlike the usual phonon case this cross section depends on the magnetic form factor, the $(1-|\hat{\kappa}_Q|^2)$ factor, and on the magnetization $\langle J^z(N\mathbf{Q}_{\mathrm{FAD}})\rangle_T^2$. Also unlike the phonon case, where the wave vector $\mathbf{q}$ is measured from a reciprocal lattice point, the wave vector $\mathbf{q}$ for magnetovibrational scattering can be measured from the magnetic satellites.

The low-temperature ($T=5$ K) excitation spectra for Tm at all wave vectors that we studied consisted of three peaks. This can be seen in Fig. 1, which shows our unpolarized neutron constant-$\boldsymbol{\kappa}$ scans at $\boldsymbol{\kappa}=(1,1,\zeta)$ 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 Fig. 1 but the two lower-energy excitations were weaker. These spectra are also in qualitative agreement with those reported by MS, except that no evidence of the excitation at 15 meV reported by these authors was seen in our scans.

In all cases, the two lower-energy excitations show a significant dispersion. The highest-energy excitation exhibits only a weak dispersion (between 8.3 and 9.6 meV) and a linewidth that is wave-vector dependent. In Fig. 2, all the excitation energies identified from scans along the $(1,1,\zeta)$ and $(0,0,2+\zeta)$ directions have been plotted versus ζ. In this figure we have also plotted (in solid lines) the expected dispersion relation for the TA phonon (branch originating from $\zeta=0$) as well as the dispersion relations for magnetovibrational scattering from the TA phonon originating 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 phonon dispersion shown in this figure is that for Tb (of comparable mass and same structure) reported by Houmann and Nicklow.[9] The striking agreement of the lower-energy excitations and the solid lines strongly suggests that these should be identified as a

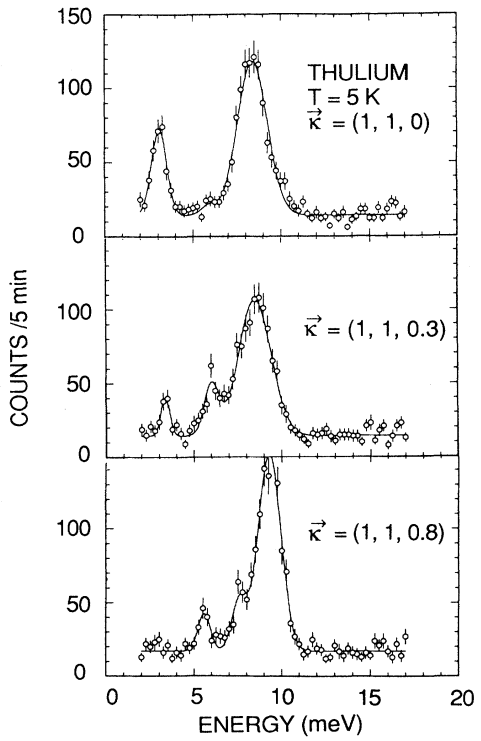

FIG. 1. Constant-$\boldsymbol{\kappa}$ scans at $\boldsymbol{\kappa}=(1,1,\zeta)$ ($\zeta=0,0.3,0.8$) for Tm at $T=5$ K. In all cases three excitations were observed: the lower-energy ones show significant dispersion, while the higher-energy one exhibits only a weak dispersion. The solid lines are from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

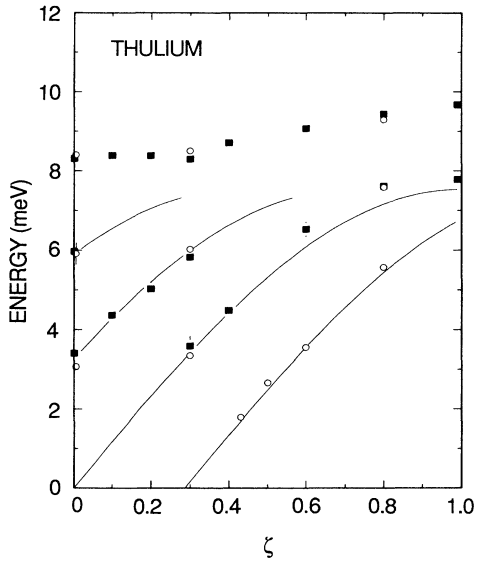

FIG. 2. Dispersion relations of the observed excitations in Tm along the $(1,1,\zeta)$ (open circles) and $((0,0,2+\zeta)$ (full squares) directions. The solid lines correspond to the dispersion relation expected for the creation of a TA phonon through nuclear as well as magnetic interactions (magnetovibrational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

TA phonon and magnetovibrational scattering from the TA phonon. The temperature dependence of these excitations, observed by MS[2] and ourselves, is also consistent with this interpretation: the intensity of those associated with magnetovibrational scattering decreases at higher temperatures and vanishes at the Néel temperature, as required by the $\langle J^z(NQ_{\rm FAD})\rangle_T^2$ dependence of Eq. (2).

We must remark, however, that while we observed these excitations in both $(1,1,\zeta)$ and $(0,0,2+\zeta)$ scans, magnetovibrational scattering from a phonon should not be observed along the c axis due to the $(1-|\hat{\kappa}_Q|^2)$ dependence of the cross section of Eq. (2). We attribute the observation of magnetovibrational scattering in scans at $(0,0,2+\zeta)$ to multiple scattering processes, i.e., the creation of a phonon followed or preceded by Bragg scattering.[10] A scattering event where the TA phonon cross section and the $(1-|\hat{\kappa}_Q|^2)$ factor are both favorable may then be shifted by a Bragg vector to the c^* axis. We have found that the probability of such a process is considerably increased in the FAD phase of thulium due to the presence of numerous magnetic satellites. In order to verify that the lower-energy excitations along the $(0,0,2+\zeta)$ direction arose from multiple scattering, we changed the fixed final neutron energy to $E_f=11.58$ meV and repeated the scan at (0,0,2). As a result, the lower-energy peaks virtually disappeared and only the mode at ≈ 8.3 meV persisted, confirming that the low-energy excitations are indeed caused by multiple scattering.

The inelastic neutron-scattering measurements using polarized neutrons and full polarization analysis were performed in order to confirm our previous claim that the highest-energy mode of Fig. 1 is magnetic and corresponds to transverse (spin-wave) excitations,[3] and that the lower modes correspond to a TA phonon or to magnetovibrational scattering from a TA phonon. These measurements were performed at constant $\kappa=(1,1,0)$ with a polarizing magnetic field of 0.5 T along the c axis. In this geometry the spin-flip (SF) scattering consists of one-half of the transverse magnetic cross section, while the non-spin-flip (NSF) scattering contains the longitudinal or nonmagnetic processes.[11] Note that the longitudinal NSF cross section contains the magnetovibrational scattering since the phonons couple through $\langle J^z\rangle$.

The results of polarized neutron measurements are shown in Fig. 3, where the open and solid circles denote the observed NSF and SF intensities. Because these measurements were performed with relaxed energy resolution, in order to obtain higher intensity, in this scan only two excitations (at 3.3 and 8.4 meV) were resolved. It is clear that the two observed excitations are of different character. The lower-energy ($E=3.3$ meV) excitation is NSF scattering, while the highest-energy excitation ($E=8.4$ meV) is predominantly (by a factor of 2) in the SF channel. The apparent spin-flipping ratios at both excitation energies are considerably smaller than the ratio observed at the (1,1,0) and $(1,1,0)+\mathbf{Q}_{\rm FAD}$ Bragg reflections (where the NSF to SF ratio is 10 ± 1). This observation may indicate that these excitations have a mixed (longitudinal and transverse) character. However, we believe that these apparent poor flipping ratios are most likely due to multiple scattering effects that artificially enhance the NSF and SF cross sections because they introduce the scattering geometries of other vectors related by a reciprocal lattice vector. To a lesser extent these poor flipping ratios could also be attributed to degradation of the spin-flipper efficiency at positions away from where it was tuned. We therefore conclude that the higher-energy excitation is of purely SF (spin-wave) character, and that the lower-energy excitation is of purely NSF character, as expected from magnetovibrational scattering.

In summary, our measurements confirm that only the higher-energy mode shown in Fig. 2 corrresponds to transverse magnetic excitations. The lower-energy excitations are of magnetovibrational origin as discussed

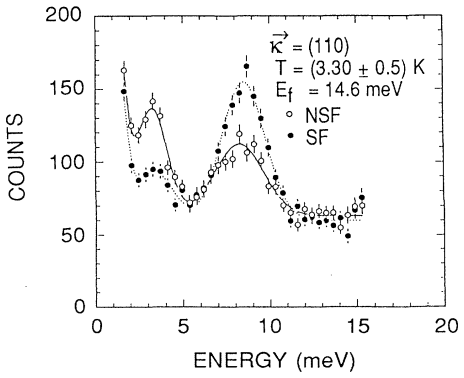

FIG. 3. Constant-κ scan at $\kappa=(1,1,0)$ showing the neutron polarization dependence of the scattering. The open circles correspond to the non-spin-flip (NSF) scattering, while the solid circles correspond to spin-flip (SF) scattering. The lower-energy excitation is NSF, while the higher-energy excitation in SF.

above. These excitations follow the TA phonon dispersion relation, the temperature dependence predicted by Eq. (2), and are of NSF character. The observation of these excitations in scans along the c axis, which is forbidden according to the cross section of Eq. (2), has been found to arise from multiple scattering processes. No evidence of the excitation at 15 meV reported by MS[2] was found. It has been recently suggested that this excitation might correspond to a crystal-field level caused by the presence of hydrogen in Tm.[12]

The small dispersion of the magnetic 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 transiton in the exchange-perturbed crystal-field level scheme. The energy of this transition can be estimated using the crystal-field parameters deduced by Touborg[13] from paramagnetic susceptibility measurements on dilute Tm alloys and pure Tm. This value is 6.7 meV (from the dilute alloys estimation) or 7.0 meV (from the pure Tm estimation). Because these excitations are "magnons" (or magnonlike), the cross section of Eq. (1) suggests that several branches of the dispersion relation should be observed at a given κ vector. If such branches exist we have not been able to resolve them due to our relatively coarse energy resolution ($\Delta E_{\mathrm{FWHM}} = 1.4$ meV at 8 meV in our unpolarized neutron measurements). However, the fact that these excitations are broader than the energy resolution and that their linewidths are wave-vector dependent is consistent with a flat magnon branch which is sharp but translated to wave vectors $\pm N\mathbf{Q}_{\mathrm{FAD}}$ and hence show an apparent maximum width of 2 meV.

Note added in proof. After this paper was submitted for publication we learned of new work performed by K. A. McEwen, U. Steigenberger, and J. Jensen. In contrast with Ref. 2 the new measurements of these authors agree with the results presented in this paper.

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