Magnetic excitations in the longitudinally modulated spin structure of erbium metal

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The magnetic excitation spectrum for the longitudinally modulated incommensurate spin structure of Er metal has been studied by inelastic neutron scattering. Since the Er spins order parallel to the *c* axis of the hcp crystal structure, it is possible to separate the longitudinal and transverse spin fluctuations. We find no sharp spin waves for the transverse fluctuations owing to the spatial variations of the exchange and anisotropy energies. The longitudinal fluctuations, however, produce neutron scattering, at small wave vectors, which possesses rather sharp peaks that have a linear dispersion relation.

While all the heavy rare-earth metals from Tb to Tm possess quite unusual and complex long-range magnetic ordering, the magnetic phases of Er are perhaps the most unusual.[1] Below about 18 K, the magnetic structure of Er (which has the hcp crystal structure) is the so-called ferromagnetic spiral or conical structure. In this structure the directions of the localized atomic moments are a superposition of a ferromagnetic component parallel to the *c* axis and a basal-plane component that rotates from plane to plane, forming a spiral with a wave vector, $\vec{q}_0$, that is incommensurate with the crystal lattice. Just above 18 K the *c*-axis component abruptly changes from a ferromagnetic ordering to a complicated type of antiferromagnetic ordering corresponding to an antiphase domain configuration with four moments up followed by four moments down. As the temperature is raised the *c*-axis modulation gradually becomes rounded and the magnitude of the ordered basal-plane moment decreases. Above 52 K (up to the Néel temperature of 84 K) the basal-plane component is not ordered at all, and the *c*-axis component is modulated nearly sinusoidally, giving a longitudinally modulated spin structure.

In the conical structure the total localized moment on each ion participates in the long-range magnetic order. In this case a theoretical description of the magnetic excitations in terms of spin waves is relatively straightforward,[2] although the interpretation of a detailed comparison between previous neutron measurements[3] and theory[2] has required that a careful theoretical treatment of the large single-ion anisotropy of Er be carried out.[4,5]

In the longitudinally modulated structure only a portion (which varies from atom to atom) of the total localized moment participates in the long-range order. The types of magnetic excitations to be expected in such a structure and their relationship to the interatomic exchange and anisotropy interactions are not yet clear from a theoretical point of view. As shown by Cooper *et al.*,[2] the equations of motion of the spins are very complicated, and they do not have solutions corresponding to the usual type of spin-wave excitations. More recently, Liu has reexamined this problem.[6] He has shown that the transverse magnetic response should be broad owing to the spatial variations of the anisotropy and exchange energies, and it may show dispersion if the anisotropy field is small compared to the exchange field. Other types of collective excitations related, perhaps, to the time-dependent fluctuations of the phase and amplitude of the moment modulations, as envisaged for charge-density wave systems,[7] have not been theoretically investigated to our knowledge. Consequently, we have carried out a neutron scattering investigation in order to characterize experimentally the magnetic excitation spectrum for Er which possesses one of the few longitudinally modulated spin-density wave structures in nature. Similar work has been carried out by Fincher *et al.*[8] on the transverse spin-density wave structure of Cr.

The major portion of the measurements was carried out between 60 and 75 K. At these temperatures the modulation wave vector $\vec{q}_0 \cong (0,0,\zeta_0)2\pi/c$ with $\zeta_0 \approx 0.29$. Two main experimental scattering configurations were employed: one with the neutron scattering vector $\vec{Q}$ parallel to the *c* axis, and hence parallel to the ordered moment direction, and one with $\vec{Q}$ nearly perpendicular to *c*. Since the elastic diffraction cross section depends on the static moment component that is perpendicular to $\vec{Q}$, the "Bragg" diffraction peaks for this longitudinally modulated structure occur only at $(h,k,l\pm\zeta_0)$ for *h* and/or $k \neq 0$, where *h*, *k*, and *l* are the usually allowed Miller indices for the hcp structure. The wave vector $\vec{q}$ of a magnetic excitation is measured relative to the positions of these satellites even for $h=k=0$. Since the inelastic scattering cross section depends on only the moment fluctuations perpendicular to $\vec{Q}$, only transverse fluctuations are measured when $\vec{Q} \parallel c$ while both transverse and longitudinal fluctuations are measured when $\vec{Q} \perp c$.

For each scattering configuration the measurements can be catagorized into two groups. In one group energy scans for constant $\vec{Q}$ with $\vec{Q}$, in reduced units, between (002) and (003), between (100) and (101), and between (110) and (111), show rather broad intensity distributions having some structure that depends on $\vec{Q}$ but no sharp peaks. Some typical results for such measurements are shown in Fig. 1. For all $\vec{Q}$ significant intensity is observed from zero energy up to energies slightly above that ($\sim$4 meV) corresponding to the maximum spin-wave energy observed in the low-temperature conical phase,[3] e.g., the peak at $\vec{Q}=(0,0,3)$ at 10 K in Fig. 1. These measurements show that no well-defined magnons, i.e., propagating transverse moment fluctuations, with large $\vec{q}$ exist in the spin-density wave structure of Er. This result was not unexpected. Theoretical work had led to the same conclusion.[2,6] Since in an incommensurate longitudinally modulated magnetic structure such as this, all possible values of the magnetic moment between zero and the maximum localized moment exist, then all possible values of exchange and anisotropy interaction magnitudes are sampled by the various atoms, leading to a continuum of possible magnetic excitation energies for the crystal.

The second group of measurements was obtained in $\vec{Q}$ scans with the energy transfer held constant at various values between 0.1 and 0.5 THz. In these measurements $\vec{Q}$ was scanned through values that included the positions of magnetic satellites, $\vec{\tau}\pm\vec{q}_0$, where $\vec{\tau}$ is a reciprocal-lattice vector of the nuclear lattice. Because of the results shown in Fig. 1, and because of the complexity of the spin-wave dispersion relation at large $\vec{q}$ for the conical phase,[3] we believed such scans would be the most likely to provide unambiguous results concerning the possible existence of well-defined acoustic magnons at small $\vec{q}$. Results for $\vec{Q} \parallel c$ near $(0,0,2-\zeta_0)$ are shown in Fig. 2 and those for $\vec{Q} \perp c$ near $(1,1,-\zeta_0)$ are shown in Fig. 3. In both cases results are shown for the energy transfer of 0.3 THz. Since $|\vec{q}_0|$ is temperature dependent, we have plotted $\vec{q}$ relative to the magnetic satellite positions so that data for different temperatures can be compared more easily. At 10 K, for both $\vec{Q} \parallel c$ and $\vec{Q} \perp c$, two well-defined, resolution broadened, peaks are observed that correspond to the acoustic spin waves at $\vec{q}_0\pm\vec{q}$ as we had measured previously.[3] At this temperature the conical structure produces intense satellite diffraction peaks near both (002) and (110) due to the spiral-type ordering of the basal plane component of the atomic moments. The results for $\vec{Q} \perp c$ appear to be more complicated than those for $\vec{Q} \parallel c$ because of the additional phonon scattering which is observed and because the focussing property of the spectrometer resolution strongly influences the peak shapes for $\vec{q} \perp \vec{Q}$, producing in this case a broad peak for $q < q_0$ and a sharp peak for $q > q_0$.

At higher temperatures ($T > 60$ K) in the longitudinally modulated phase, the scattering for $\vec{Q} \parallel c$ and for $\vec{Q} \perp c$ are very different. For $\vec{Q} \parallel c$ the intensity distribution has a peak centered at $\vec{\tau}\pm\vec{q}_0$. Even with the best energy resolution employed in this study ($\sim$0.05 THz), we were unable to measure any structure in this peak. Consequently, we believe it does not represent scattering by well-defined acoustic spin-wave modes with a very steep dispersion relation. However, as shown in Fig. 4, the q width increases with increasing energy up to an energy of

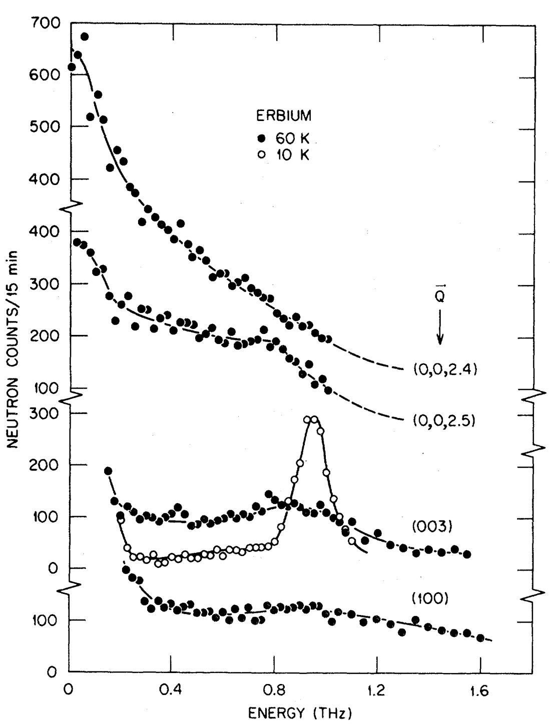

FIG. 1. Constant-Q measurements of the energy distribution of neutrons scattered by Er metal at 60 and 10 K.

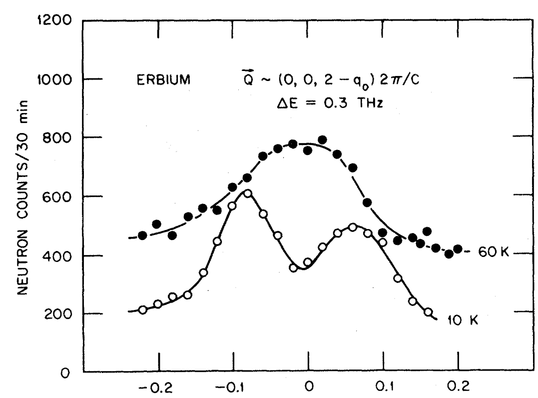

FIG. 2. Constant-ΔE measurements of the neutron scattering for $\vec{Q} \parallel c$ near $\vec{Q}=(0,0,2-\zeta_0)$ at several temperatures.

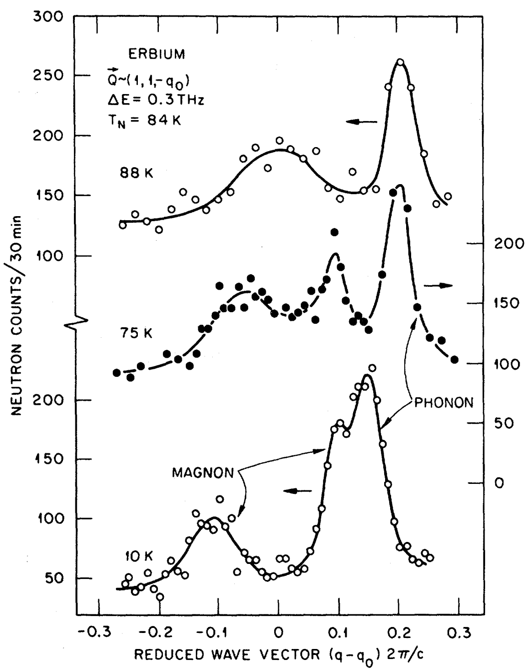

FIG. 3. Constant-ΔE measurements of the neutron scattering for $\vec{Q}\perp c$ near $\vec{Q}=(1,1,-\zeta_0)$ at several temperatures.

about 0.5 THz, above which the intensity becomes too small to measure. Thus the scattering by transverse magnetic fluctuations appears to be a cone of intensity, positioned at the wave vector of the modulated structure, in addition to the broad scattering illustrated in Fig. 1. In sharp contrast to these

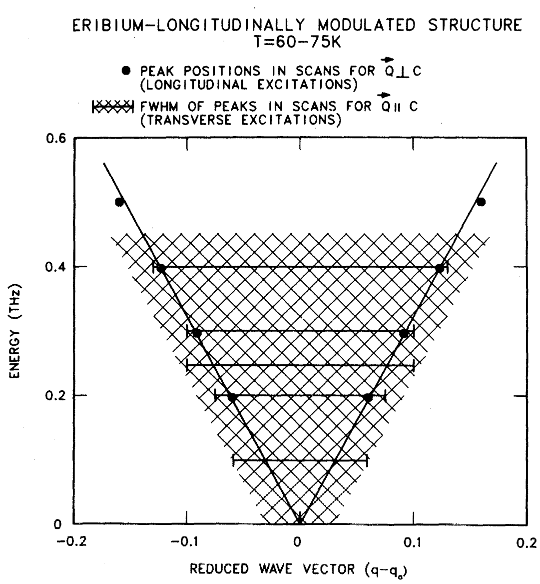

FIG. 4. Magnetic excitation spectra measured for Er metal in the longitudinally modulated structure. The straight lines shown are to guide the eye and do not represent a theoretical calculation.

results those for $\vec{Q}\perp c$ show two rather well-defined peaks displaced in $\vec{q}$ from the satellite position. They are still observed even at $T=75$ K, as shown by the middle scan of Fig. 3. Because $|\vec{q}_0|$ is temperature dependent, the relative positions of the magnon and phonon scattering change with temperature. Since these two peaks are seen only for $\vec{Q}\perp c$, we believe they represent some type of propagating longitudinal excitation. Of course, we should also see scattering from the transverse fluctuations. From the measurements with $\vec{Q}\parallel c$, we expect the transverse fluctuations to give a peak at $\vec{q}_0$ as seen in Fig. 2. The results shown in Fig. 3 are not inconsistent with a superposition of a broad peak centered at $\vec{q}_0$ and two sharper peaks displaced parallel to the c axis on either side of $\vec{q}_0$. From the measurements shown in Figs. 2 and 3 and others carried out for different ΔE and near different satellites we obtained the results shown in Fig. 4. The dispersion relation for the longitudinal excitations appears to be linear at least up to about 0.5 THz. Above 0.5 THz the intensity becomes too small to obtain results in a reasonable length of time. The dispersion relation is temperature independent, at least for 60 K $< T < T_N$, and it is essentially the same as the dispersion relation observed for the spin waves in the conical phase at 5 K.[3] Also, it seems to coincide closely with the $(\vec{q},E)$ boundary defined by the full width at half maximum (FWHM) of the scattering measured with $\vec{Q}\parallel c$ for the transverse fluctuations (see Fig. 4) above 60 K.

The close relationship between the small-$\vec{q}$ longitudinal and transverse excitation energies suggests that both types of excitations are related similarly to the exchange and crystal-field interactions. Presumably the broad "cone" of scattering for the transverse excitations is indicative of large damping for these modes. However, it is surprising then that the damping appears to be so unsymmetric, so as to produce, for a given $\vec{q}$, much more intensity on the high-energy side of the low-temperature dispersion relation than on the low-energy side. Just the opposite behavior (peaks having low-energy tails) seems to occur for the high q excitations, and in some ferromagnets.[9,10]

Also shown in Fig. 3 are the results obtained for $\vec{Q}\perp c$ and $T=88$ K $> T_N$. Similar results are obtained for $\vec{Q}\parallel c$ at this temperature. Slightly above T_N the scattering, as measured in such $\vec{Q}$ scans, continues to possess a peak at $\vec{q}_0$, but no evidence for magnetic peaks at other wave vectors, and no differences between measurements for $\vec{Q}\parallel c$ and $\vec{Q}\perp c$ are seen.

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