

## Magnetic Excitations in Commensurable Periodic Structures

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The magnetic excitations of a  $\text{Ho}_{90}\text{Tb}_{10}$  single crystal have been studied by inelastic neutron scattering. In the commensurable helical structure the bunching of the moments about the easy directions of magnetization induces an energy gap at long wavelengths whose magnitude, compared to that in the ferromagnetic phase, implies an anisotropic two-ion coupling. The strong dipolar forces give rise to a discontinuity in the dispersion relations at  $\mathbf{q}=\mathbf{0}$ , and resolve the longstanding mystery of the stability of the conical structure in Ho at low temperatures.

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Two decades ago Koehler *et al.*<sup>1</sup> showed by neutron diffraction that, at low temperatures, Ho may form a magnetic structure which is commensurable with the lattice periodicity. The moments on the atoms in a particular plane normal to the  $c$  axis of the hcp structure are all parallel, but such planes of moments bunch in pairs successively about the six magnetically easy  $b$  directions, thus repeating periodically every twelve atomic layers. Recently Gibbs *et al.*<sup>2</sup> have shown by x-ray diffraction that such pairs of bunched planes also occur at higher temperatures, with "spin slips" of single planes. The study of magnetic excitations in rare-earth metals has provided a wealth of information which is not available from the magnetic structures alone, and we therefore decided to study the excitations in such a commensurable periodic structure.

Rather than using pure Ho, whose stable low-temperature structure is shallow cone, we performed our experiments on a crystal containing 10% of Tb, which has the desirable effects of confining the moments to the basal plane, and inducing the simple commensurable helical structure below 30 K and ferromagnetism below 20 K. Earlier studies in this ferromagnetic phase<sup>3</sup> revealed an energy gap in the spin-wave spectrum at long wavelengths. In the incommensurable helical structures of the heavy rare earths this energy gap vanishes, essentially because of the breaking of the translational symmetry in the  $c$  direction. In the commensurable structure, the symmetry is not broken, but merely modified, and there is no requirement that the energy gap vanish.

The spin-wave energies in this phase, measured with conventional inelastic neutron-scattering techniques at the DR3 reactor at Risø and shown in Fig. 1, do indeed exhibit such an energy gap. These results contain a

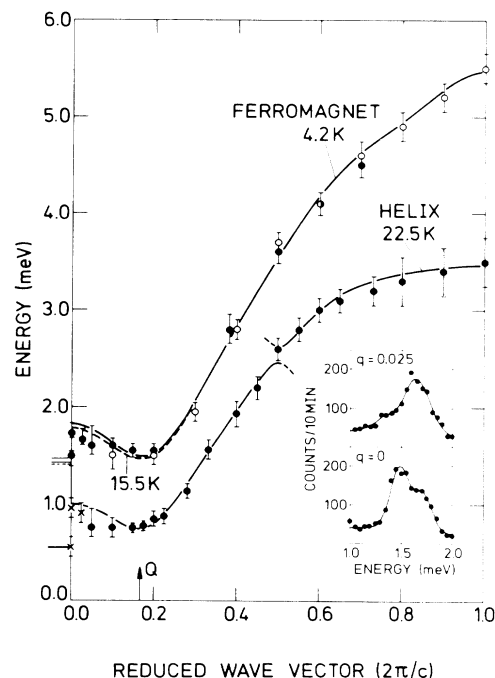


FIG. 1. Dispersion relations for magnetic excitations propagating in the  $c$  direction in  $\text{Ho}_{90}\text{Tb}_{10}$ . Upper branch: Ferromagnetic phase. The open symbols are measurements at 4.2 K (Ref. 3); the filled symbols are our results at 15.5 K; and the full and dashed lines show the theoretical dispersion relations at 4.2 and 15.5 K, respectively. The calculated long-wavelength energies in the basal plane are shown to the left of the ordinate axis and the discontinuity, which is due to the dipolar interaction, is clearly manifested in the experimental measurements (inset). Lower full curve: Calculated dispersion relation for the commensurable helical structure.

number of features whose elucidation requires a detailed consideration of both the magnetic structures and the excitations. These have been calculated with a self-consistent molecular-field method, combined with the random-phase approximation for the excitations. The starting point is the Hamiltonian

$$H = \sum_{ilm} B_l^m O_l^m(\mathbf{J}_i) - \frac{1}{2} \sum_{ij} \sum_{\alpha\beta} \mathcal{J}^{\alpha\beta}(ij) J_{\alpha i} J_{\beta j}.$$

The first term is the single-ion crystal-field contribution, involving the Stevens operators  $O_l^m(\mathbf{J})$ . Because of the magnitudes of the relevant multipoles of the  $4f$  charge clouds,  $B_2^0$  in Ho is relatively small while  $B_6^0$  and  $B_6^6$  are very large. In comparison with the very large hexagonal anisotropy, the magnetoelastic  $\gamma$  strains, which have a large influence on the magnons<sup>4</sup> in Tb, may be neglected in Ho. In Ho  $B_2^0$  is positive but  $B_6^0$  is negative, and the rapid increase of  $\langle O_6^0 \rangle$  at low temperatures tilts the moments out of the plane. We take the two-ion coupling terms to comprise an isotropic Heisenberg exchange and the dipolar coupling between the moments, so that the Fourier transform is

$$\mathcal{J}^{\alpha\beta}(\mathbf{q}) = \mathcal{J}(\mathbf{q}) \delta_{\alpha\beta} + (g^2 \mu_B^2 / V) D^{\alpha\beta}(\mathbf{q}),$$

where  $V$  is the volume per atom. The dipolar contribution<sup>5</sup> is highly anisotropic and discontinuous at  $\mathbf{q} = \mathbf{0}$ . For  $\mathbf{q}$  along the hexagonal  $c$  axis it is diagonal with components, in the long-wavelength limit,

$$D^{cc}(\mathbf{q}) = -8\pi/3; \quad D^{bb}(\mathbf{q}) = D^{aa}(\mathbf{q}) = 4\pi/3.$$

In addition, since the  $c/a$  ratio is not ideal, the dipolar coupling gives a small effective contribution to  $B_2^0$ .

At 4.2 K the magnetic structure of Ho is distorted, shallow cone, with a small ordered moment in the  $c$  direction and a bunched helical structure in the plane described by

$$\langle J_a(i) \rangle = \langle J_p \rangle (u \sin \mathbf{Q} \cdot \mathbf{R}_i - v \sin 5\mathbf{Q} \cdot \mathbf{R}_i),$$

$$\langle J_b(i) \rangle = \langle J_p \rangle (u \cos \mathbf{Q} \cdot \mathbf{R}_i + v \cos 5\mathbf{Q} \cdot \mathbf{R}_i),$$

where  $\mathbf{Q}$  is a vector of magnitude  $\pi/3c$  in the  $c$  direction and

$$u \equiv \cos(\pi/12 - \phi); \quad v \equiv \sin(\pi/12 - \phi).$$

$\phi$  is the angle in the plane between any moment and the closest easy axis of magnetization, as illustrated in Fig. 2. The magnetization for magnetic fields in the  $c$  and  $a$  directions was calculated by the self-consistent molecular-field method applied to the  $J=8$  ions, with adjustment of the crystal-field parameters to fit experimental data by various authors.<sup>1,6</sup> The absolute value of  $\mathcal{J}^{bb}(\mathbf{Q}) = 0.475$  meV was estimated from the Néel temperature, while  $\mathcal{J}^{bb}(5\mathbf{Q})$  and  $\mathcal{J}^{cc}(\mathbf{0})$  were extracted from the excitation energies in the helical phase. The fit to the data is shown in Fig. 2 and the resulting parameters given in Table I. The best fit was obtained with a small

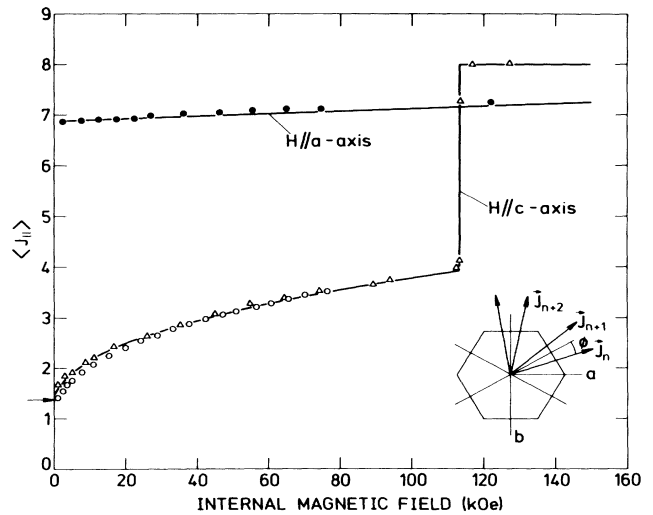


FIG. 2. Calculated  $4f$  contribution to the magnetization of Ho at 4.2 K (given by  $g\mu_B\langle J_{11} \rangle$  per ion), compared with experimental values deduced from Ref. 6. The zero-field structure is a bunched cone, comprising the illustrated bunched helical structure in the plane, and a small moment in the  $c$  direction. The value of the  $c$ -axis moment, deduced from neutron-diffraction measurements in Ref. 1, is indicated by the arrow.

$B_4^0$  ( $|B_4^0| \lesssim 10^{-5}$  meV), and a value of  $B_6^0$  very close to the point-charge value  $-77B_6^0/8$ . The hexagonal anisotropy is only approximately determined by the  $a$ -axis magnetization, but much more precisely by the bunching angle  $\phi$ , also measured by neutron diffraction.<sup>1</sup> Our calculations also account well for the cone angle as a function of temperature.

The magnetic-excitation spectrum may be determined from the poles of the generalized susceptibility tensor, which in the random-phase approximation is calculated in terms of  $\mathcal{J}^{\alpha\beta}(\mathbf{q})$  and  $\bar{\chi}(\omega)$ , the frequency-dependent susceptibility of an ion in the molecular-field approximation. For the alloy we used the virtual-crystal approximation, assuming the exchange coupling to be proportional to the  $g-1$  factors of the ions involved. For  $\bar{\chi}_{Tb}(\omega)$  we used the spin-wave approximation, with parameters determined from earlier measurements.<sup>4</sup> The introduction of the Tb ions increases the effective axial anisotropy, so that the planar helix undergoes a first-order transition to a ferromagnetic phase before becoming unstable to the formation of a cone structure, in accordance with the experimental observations. In order to obtain the correct energy gap in the bunched helix at 22.5 K, it was necessary to decrease  $B_2^0$  for the Ho ions by about 20% relative to the value in the pure metal, an effect which may be ascribed primarily to the influence of the alloying on the ionic environment, including the local strains. As may be seen in Table I, this model also gives good agreement between the calculated and measured bunching angles at two temperatures. These were

determined either from the ratio of the intensity of the fifth to the first harmonic of the magnetic reflections or, the total ordered moment known, by comparison of the intensities of magnetic and nuclear reflections. These two methods gave consistent results<sup>7</sup> for  $\phi$ .

In the spin-wave approximation, the excitation energies in the bunched helical phase in the  $c$  direction are given by

$$\begin{aligned} \epsilon^2(\mathbf{q}) = & [A+B+J\{\mathcal{J}^{cc}(\mathbf{0}) - \mathcal{J}^{cc}(\mathbf{q})\}](A-B+J\{u^2[\mathcal{J}^{bb}(\mathbf{Q}) - \frac{1}{2}\mathcal{J}^{bb}(\mathbf{q}+\mathbf{Q}) - \frac{1}{2}\mathcal{J}^{bb}(\mathbf{q}-\mathbf{Q})] \\ & + v^2[\mathcal{J}^{bb}(5\mathbf{Q}) - \frac{1}{2}\mathcal{J}^{bb}(\mathbf{q}+5\mathbf{Q}) - \frac{1}{2}\mathcal{J}^{bb}(\mathbf{q}-5\mathbf{Q})\}]), \quad (1) \end{aligned}$$

where

$$A+B = 6J_1B_2^0 - 60J_3B_4^0 + 210J_5B_6^0 + 6J_5B_6^0 \cos 6\phi + J[u^2\mathcal{J}^{bb}(\mathbf{Q}) + v^2\mathcal{J}^{bb}(5\mathbf{Q}) - \mathcal{J}^{cc}(\mathbf{Q})],$$

$$A-B = 36J_5B_6^0 \cos 6\phi; \quad J_n \equiv (J - \frac{1}{2}) \cdots (J - n/2).$$

For an infinitesimal  $\mathbf{q}$  in the  $c$  direction, the square of the energy gap  $\Delta_c^2$  is given by  $(A+B+4\pi g\mu_B M)(A-B)$ , where  $M$  is the magnetic moment per unit volume, the experimental value of which corresponds to  $4\pi g\mu_B M = 0.28$  meV. A similar analysis for  $\mathbf{q}$  in the basal plane gives  $\Delta_b^2 = (A+B)(A-B)$ . Since the total axial anisotropy  $A+B$  is small, the magnon energies in the  $c$  direction are increased substantially by the dipolar coupling, while those in the basal plane are unaffected. The discontinuity at  $\mathbf{q}=\mathbf{0}$  occurs over a range of the order of ten times the inverse of the sample dimensions.<sup>5</sup>

The discontinuity at  $\mathbf{q}=\mathbf{0}$  is clearly seen in the ferromagnetic phase, as illustrated in the inset to Fig. 1. The discontinuity in the helical phase is not experimentally as clearly resolved because of the background from the magnetic Bragg scattering. However, the inelastic magnetic scattering is consistent with a two-peak structure similar to that in the ferromagnet. The energy gap is considerably lower than in the ferromagnetic phase, which is at first sight surprising since, according to (1), it should only be reduced by a factor of about  $\cos 6\phi$  or roughly 0.8. The discrepancy can be accounted for by a decrease of  $B_2^0$  by about 50% in the helical phase, as indicated in Table I. Such an effect could be produced by an anisotropic two-ion coupling of the type previously observed<sup>4</sup> in Tb, which has a different influence on the microscopic and macroscopic properties in the two phases. Specifically the term  $\mathcal{C}(\mathbf{q})$ , defined in Ref. 4, which is

TABLE I. Crystal-field parameters. The values of  $B_2^0$  used in the calculations of the magnetic properties of Ho and Ho<sub>90</sub>Tb<sub>10</sub> in different phases are given, together with calculated and experimental bunching angles. The remaining crystal-field parameters are as follows:  $B_4^0 = 0$ ,  $B_6^0 = -0.956 \times 10^{-6}$  meV,  $B_8^0 = 9.21 \times 10^{-6}$  meV.

Phase	Temp. (K)	$B_2^0$ (meV)	$\phi_{\text{calc.}}$ (deg)	$\phi_{\text{meas.}}$ (deg)
Ho (cone)	4.2	0.024	5.81	5.8
Ho <sub>90</sub> Tb <sub>10</sub> (ferro.)	4.2	0.031	0	0
Ho <sub>90</sub> Tb <sub>10</sub> (helix)	22.1	0.019	7.81	$7.4 \pm 0.8$
Ho <sub>90</sub> Tb <sub>10</sub> (helix)	28.1	0.019	9.15	$8.7 \pm 0.8$

known to be substantial in Tb, gives a contribution  $\mathcal{C}(\mathbf{0})$  to  $A+B$  in the ferromagnetic phase and  $\mathcal{C}(3\mathbf{Q}) \cos 6\phi$  in the helical phase. A more precise elucidation of the nature of the anisotropic two-ion coupling would require further measurements in a magnetic field.

The full lines in Fig. 1 are the dispersion relations calculated with the parameter of Table I and the exchange functions of Fig. 3. The full RPA theory was used in these calculations, rather than the spin-wave approximation. The latter works satisfactorily at low temperatures

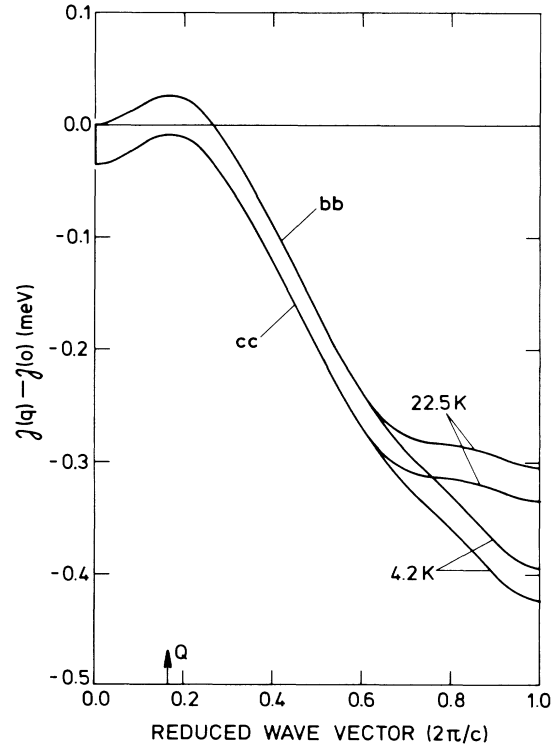


FIG. 3. Parallel and perpendicular components of the Fourier transform of the two-ion coupling between Ho ions, when  $\mathbf{q}$  is along the  $c$  direction. The discontinuity at  $\mathbf{q}=\mathbf{0}$  in the parallel component is due to the dipolar coupling, which also stabilizes the cone structure in Ho at low temperatures.

but the crystal-field parameters are renormalized. The calculated energy gap halfway to the zone boundary in the commensurate helical phase is due to the effective doubling of the magnetic unit cell in the helical coordinate system by the bunching, which is neglected in Eq. (1). The two modes correspond to an in-phase and an out-of-phase oscillation of the paired moments, but the energy difference between them is too small to resolve with the peak widths of almost 1 meV which are observed in the vicinity of the gap.<sup>7</sup> Except for a modification near the zone boundary, we have used the same  $\mathcal{J}(\mathbf{q})$  in the ferromagnetic and helical phases of the alloy.

We have calculated the dispersion relations in the bunched conical and helical phases of Ho and find reasonable agreement with the measurements of Stringfellow *et al.*<sup>8</sup> and Nicklow.<sup>8</sup> Whenever the moments are not along directions of high symmetry,  $B_2^0$  mixes the longitudinal, zero-energy response with the transverse excitations. Beyond the RPA this mixing would reduce the energy of the long-wavelength excitations at 22.5 K by as much as about 0.1 meV, which would be compensated by an increase of  $B_2^0$  by about 10%. The importance of the mixing increases with temperature, and  $B_2^0$  substantially modifies the excitation spectrum<sup>8</sup> at 50 K and above.

The peak in the basal-plane component of  $\mathcal{J}(\mathbf{q})$  in Fig. 3 stabilizes the helical structure. The discontinuity in the  $c$ -axis component at  $\mathbf{q}=\mathbf{0}$  resolves the long-standing mystery of the stability of the cone structure of Ho at low temperatures. Elliott<sup>9</sup> and Sherrington<sup>9</sup> pointed out that, with isotropic exchange and anisotropy parameters varying smoothly with temperature, it is energetically favorable to form not a cone but a tilted helix, in which the  $z$  component of the exchange energy is lower than in the cone. As illustrated in Fig. 3, however, the increase in dipolar energy associated with the formation of a longitudinal wave forces  $\mathcal{J}^{cc}(\mathbf{Q})$  below  $\mathcal{J}^{cc}(\mathbf{0})$  in the  $c$  direction, thus prohibiting the tilted helix. Correspondingly, the mode in the helical phase which tends to go

soft when the axial anisotropy reaches a critical value is not at  $\mathbf{Q}$  but rather the lowest mode at  $\mathbf{q}=\mathbf{0}$ .

We have, in summary, observed the energy gap in the commensurate helical structure which is a consequence of the bunching of the moments. We have developed a self-consistent molecular-field theory which accounts for the magnetic structures of Ho and Ho<sub>90</sub>Tb<sub>10</sub>, including the bunching angles and, within the random-phase approximation, the dispersion relations of the excitations. The surprisingly large decrease in the energy gap between the ferromagnetic and helical phases indicates an anisotropic two-ion coupling of the type observed earlier in Tb. The previously neglected dipolar interaction plays a crucial role both in creating a discontinuity in the excitation spectrum at  $\mathbf{q}=\mathbf{0}$  and in stabilizing the cone phase in Ho at low temperatures.

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