Magnetic Anisotropy in Rare-Earth Metals

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The magnetic field dependence of the energy of long-wavelength mangons in Tb-10% Ho has been studied by inelastic neutron scattering. The results agree with the "frozen-lattice" model, provided that the second-order magnetoelastic effect is taken into account. The planar anisotropy is almost entirely the result of magnetoelastic effects. The temperature dependences of the anisotropy parameters have been deduced from the results.

There has recently been extensive discussion of the mechanisms responsible for the large magnetic anisotropies in rare-earth metals, and particularly of the way in which they affect magnon energies. Cooper[1] has considered the magnon dispersion relations for ferromagnetic hcp metals, such as Tb and Dy, in which the axial anisotropy confines the ordered moments to the basal plane. In particular, he has calculated the effect of a magnetic field on the magnon energy in the long-wavelength limit, taking into account the effect of the first-order cylindrically symmetric magnetoelastic effect in the frozen-lattice model of Turov and Shavrov,[2] in which the lattice strains are assumed to be unable to follow the spin precession in the spin wave. As we shall see, our results show that the frozen-lattice model is indeed valid, but in order to account for them in detail, it is necessary also to take into account the hexagonally symmetric second-order magnetoelastic effect.

Accordingly, we consider the Hamiltonian

$$\mathcal{H}=-\sum_{i\neq j}J(\vec{\mathbf{R}}_i-\vec{\mathbf{R}}_j)\vec{\mathbf{J}}_i\cdot\vec{\mathbf{J}}_j-\sum_i\{P_2J_{i\zeta}^2-\tfrac{1}{2}P_6^6[(J_{i\xi}+iJ_{i\eta})^6+(J_{i\xi}-iJ_{i\eta})^6]\}$$
$$-\frac{2Ac}{(J-\frac{1}{2})(J-1)(J-\frac{3}{2})}\left[\frac{\bar{\epsilon}_1^{\gamma}}{4}\sum_i(J_{i\xi}^4-6J_{i\xi}^2J_{i\eta}^2+J_{i\eta}^4)-\bar{\epsilon}_2^{\gamma}\sum_iJ_{i\xi}J_{i\eta}(J_{i\xi}^2-J_{i\eta}^2)\right]$$
$$-\frac{2Cc}{(J-\frac{1}{2})}\left[\frac{\bar{\epsilon}_1^{\gamma}}{2}\sum_i(J_{i\xi}^2-J_{i\eta}^2)+\bar{\epsilon}_2^{\gamma}\sum_i(J_{i\xi}J_{i\eta})\right]-g\beta\sum_i\vec{\mathbf{H}}\cdot\vec{\mathbf{J}}_i. \tag{1}$$

We have used generally the same notation as Cooper,[1] so that $J(\vec{\mathbf{R}})$ is the indirect exchange, $\vec{J}$ the total angular momentum on the ion, P_2 and P_6^6 represent the crystal-field anisotropy, and the $\bar{\epsilon}$ are the equilibrium strains. We have explicitly written the expressions for the magnetoelastic coupling constants in terms of the first- and second-order magnetostriction coefficients[3] C and A and the reduced elastic constant $c=c^{\gamma}/J$. We have chosen Cartesian axes such that ξ is the easy direction of magnetization in the hexagonal plane, while ζ is the hexagonal axis.

This Hamiltonian may be diagonalized[4] to yield the magnon dispersion relations. When the field is applied in the easy (ξ) direction, the magnon energy in the long-wavelength limit is given by

$$\Delta_e^2=[-2P_2J-6P_6^6J^5+2c(C^2+A^2)+3cAC+g\beta H][-36P_6^6J^5+4c(C^2+A^2)+10cAC+g\beta H]. \tag{2}$$

If a field is applied in the hard direction in the plane, the magnetization rotates so that it makes an angle φ with the hard direction, given by

$$g\beta H=2\cos\varphi(4\cos^2\varphi-1)(4\cos^2\varphi-3)(3cAC-6P_6^6J^5). \tag{3}$$

The corresponding value of the energy gap is

$$\Delta_h^2=[-2P_2J-6P_6^6J^5+2c(C^2+A^2)+3cAC+(6cAC-12P_6^6J^5)(\cos4\varphi+\cos2\varphi)][4c(C^2+A^2)$$
$$+8cAC\cos6\varphi+(3cAC-6P_6^6J^5)(-5\cos6\varphi+2(\cos4\varphi+\cos2\varphi)+1)]. \tag{4}$$

Above the critical field

$$g\beta H_c = 36(\tfrac{1}{2}cAC - P_6{}^6 J^5) \tag{5}$$

at which the magnetization turns into the hard direction, the gap is given by

$$\Delta_h{}^2 = [-2P_2 J^5 + 6P_6{}^6 J^5 + 2c(C^2+A^2) - 3cAC + g\beta H][4c(C^2+A^2) + 8cAC + g\beta(H-H_c)]. \tag{6}$$

The most striking difference between Eq. (4) and the analogous expression when the frozen-lattice model does not apply is that, in the latter case, Δ_h goes to zero at the critical field,[1] while in the frozen-lattice model the energy gap remains finite at all fields, although it has a minimum at H_c.

There are two questions of primary importance concerning the contribution of magnetoelastic effects to the magnetic anisotropy and magnon energy gap. The first is whether the frozen-lattice model is valid, so that the cylindrically symmetric magnetoelastic effect contributes to Δ, and the second concerns the relative contribution of the crystal fields and second-order magnetoelastic effect to the hexagonal anisotropy. These questions can be elucidated by studying the energy gap as a function of temperature and magnetic field. Because the anisotropy coefficients have different theoretical temperature dependences,[1,9] measurements of Δ as a function of temperature in zero field yield some information about them. By such measurements, we have earlier shown[5] that magnetoelastic effects are important in Tb-10% Ho, since the temperature dependence of the gap is consistent either with the frozen-lattice model or with a model in which the lattice is not frozen but the hexagonal anisotropy is dominated by the magnetoelastic contribution. From infrared resonance measurements, Marsh and Sievers[6] reached the same conclusion about Tb, but were, in addition, able to show that the results for Dy are not consistent with the latter hypothesis. They therefore concluded that the frozen-lattice model probably applies for both metals, since their anisotropy behavior is likely to be similar. These measurements do not separate the contributions of the different anisotropy parameters directly, however, and the interpretation therefore required assumptions about their magnitudes and temperature dependences which suffer from some uncertainty and are, indeed, to some extent inconsistent with the conclusions of this paper. As may be seen from Eqs. (2)-(6), the application of a magnetic field does allow the determination of the anisotropy coefficients explicitly at any temperature. Microwave resonance measurements in a magnetic field have resulted in different conclusions. Earlier measurements (see Ref. 1) indicated that the frozen-lattice model is not applicable, but later results of Wagner and Stanford,[7] at higher frequencies, indicated that it is.

We have therefore carried out measurements of the magnon energy gap as a function of both magnetic field and temperature by inelastic neutron scattering, using the techniques described earlier.[8] The sample was a monocrystalline sphere of Tb-10% Ho with a diameter of 6.5 mm, and an external field of up to 48 kG was applied by means of a superconducting solenoid. The results of these measurements are shown in Fig. 1, from which it may immediately be concluded that the frozen-lattice model applies, since a field large enough to rotate the magnetization into a hard direction does not reduce the gap to zero.

It may be seen from Eqs. (2), (4), and (6) that a knowledge of H_c, $\Delta_h(H_c)$, $\Delta(0)$, and $\partial\Delta^2/\partial H$, either for H in the easy direction or for fields greater than H_c in the hard direction, is adequate to determine the anisotropy parameters $-2P_2J$, $-36P_6{}^6J^5$, $18cAC$, and $2c(C^2+A^2)$ at a particular temperature. Alternatively, these parameters may be deduced from a least-squares fitting of all the experimental points, and this method has generally been used to deduce the parameters of Fig. 2 from the results of Fig. 1. The fits produced by this procedure are shown by the full lines in Fig. 1, from which it is apparent that the experimental results fit the theoretical expressions within the errors of the former. If the second-order magnetoelastic effect is neglected, which amounts to neglecting A in all expressions, no satisfactory fit can be obtained. Specifically, if $\Delta_h(H_c)$ is fitted to the experimental results, the theoretical value of $\Delta(0)$ is characteristically about 20% above the experimental value.

From Fig. 2 it is clear that the hexagonal anisotropy is dominated by the magnetoelastic effect, and the crystal-field term $P_6{}^6J^5$ is zero within the experimental error. The magnetiza-

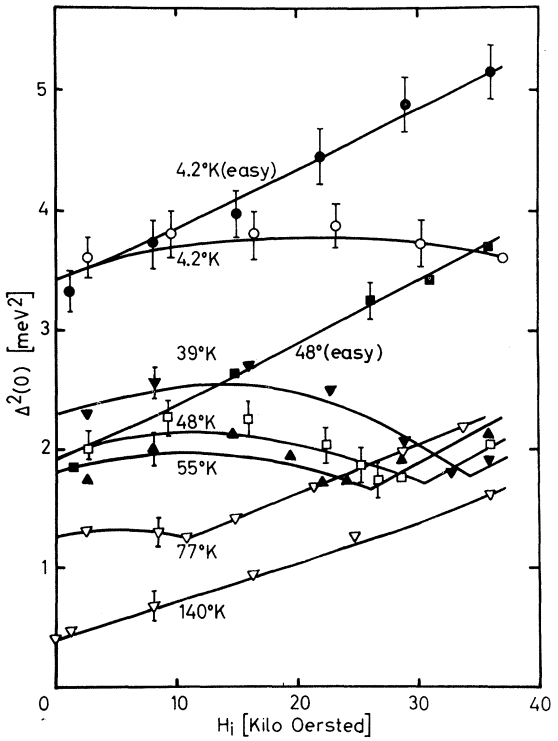

FIG. 1. The dependence of the square of the magnon energy gap in Tb-10% Ho on the internal field in the crystal at a number of different temperatures. Except where indicated, the field is applied in the magnetically hard direction in the hexagonal plane. The full lines are least-squares fits of the theoretical expressions given in the text to the experimental results.

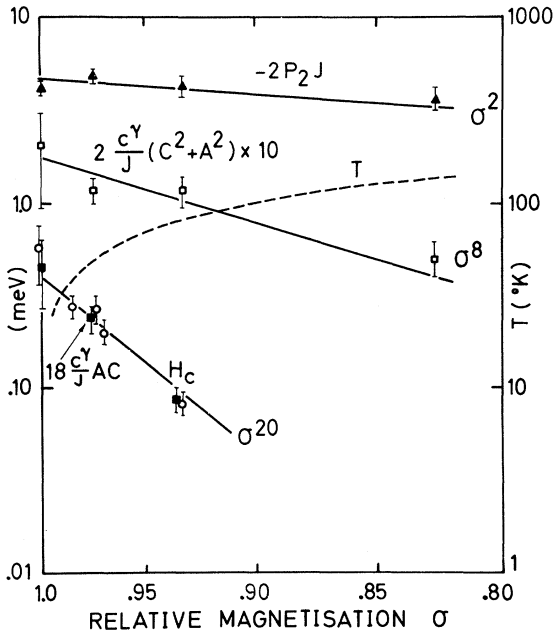

FIG. 2. The single-ion anisotropy parameters, deduced from the results of Fig. 1, as a function of magnetization plotted on a log-log scale. The dependence of the magnetization on temperature is also shown.

tion dependence of $c(C^2+A^2)$, which is dominated by the C^2 term, is consistent with either the σ^6 variation with ordered moment predicted by Callen and Callen,[9] or the σ^8 variation which Brooks[10] finds when the elliptical spin precession in the spin waves is taken into account. On the other hand, the variation of P_2 is better described by the Brooks σ^2 dependence than by the σ^3 of Callen and Callen, while that of cAC is greater than the σ^{13} predicted by Callen and Callen but agrees well with the σ^{20} variation of Brooks. It appears, therefore, that the elliptical precession of the spins may be of importance in determining the magnetization dependence of the anisotropy parameters. Using a value of 2×10^5 °K/atom for c and the C and A measurements of Rhyne and Legvold,[3] we find values of 0.14 meV/atom for $2c(C^2+A^2)$, and 0.44 meV/atom for 18 cAC. These compare satisfactorily with our experimentally determined values of 0.18 and 0.44 meV/atom, respectively, at 4.2°K.

This study of the field dependence of the energy gap has therefore allowed us to demonstrate unambiguously that the frozen-lattice model is valid, that the second-order magnetoelastic effect must be taken into account to explain the results, and that magnetoelastic effects provide the dominant contribution to the macroscopic hexagonal anisotropy. In addition, the magnetization dependences of the anisotropy parameters, which may be deduced explicitly by this method, are consistent with the theory when account is taken of elliptical spin precession,[10] and the values of the magnetostriction parameters deduced from the results agree with those measured directly. In order to obtain a monocrystalline sphere large enough to allow this experiment to be performed, it was necessary for us to use a crystal containing 10% Ho. Since the anisotropy properties of Ho are rather similar to those of Tb, we do not feel that the addition of this small amount of solute appreciably affects the single-ion parameters which we have measured. Further experiments are planned to study the magnetic anisotropy in different rare-earth metals and alloys.

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icated the results of his calculations before publication.

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Raman Light Scattering from Excitons and Magnons in Cobalt Fluoride

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We have observed one-particle, inelastic, polarized light scattering from low-lying excitons and magnons in single-crystal cobalt fluoride (CoF_2) at 2°K. Because of the orbital nature of these excitations, the cross sections (10^{-33}–10^{-32} cm^2/sr) are much larger than for one-magnon scattering in the other case, which has been measured (FeF_2, $\sim 10^{-35}$ cm^2/sr). Our measured cross sections are in good agreement with a recent calculation by Ishikawa and Moriya.

The first observation[1] of one-magnon light scattering in magnetic insulators was in FeF_2 where the magnons are predominantly spinlike excitations and the scattering mechanism requires spin-orbit coupling in the virtual intermediate state in order to be finite for an electric dipole process.[2] Subsequent measurements have concentrated on two-magnon scattering.[1,3] This is because for spinlike excitations it is a stronger process (proceeding via inter-ion exchange coupling[1,4]), and in low-anistropy materials such as manganese and nickel compounds, the one-magnon energy is only a few cm^{-1}, and hence difficult to measure. We wish to report here the observation of scattering by single magnons and low-lying excitons in CoF_2. The latter arise from excitations within the low-lying $^4T_1$ levels of the cobalt ions, split by spin-orbit coupling, orthorhombic crystal field, and inter-ion exchange interactions. The scattering mechanism involves a direct coupling of the $\vec{E}$ vector of the incident light to the orbital part of the excitations, and this gives rise to larger cross sections than for spin excitations. In fact, the exciton scattering is comparable in magnitude with the phonon scattering. Measured cross sections are in good agreement with those recently calculated by Ishikawa and Moriya.[5]

The symmetry group of CoF_2 below T_N is $D_{4h}^{14}(D_{2h}^{12})$ and branches of the excitation spectrum at $\vec{k}=0$ are labeled by unitary subgroup representations Γ_1^+, Γ_2^+, and $\Gamma_3^+ + \Gamma_4^+$. In the absence of off-diagonal inter-ion exchange coupling, all branches show the sublattice degeneracy of 2. Without this restriction the Γ_1^+, Γ_2^+ pair may show a Davydov splitting while Γ_3^+ and Γ_4^+ are required to be degenerate by the antiunitary operators of $D_{4h}^{14}(D_{2h}^{12})$. The low-lying electronic levels of the Co^{2+} ion have been studied by Johnson, Dietz, and Guggenheim[6] and Gladney.[7] The dispersion of the low-lying excitations in CoF_2 has been measured by neutron scattering,[8] and the ones we will be concerned with here are the magnons with an energy of 37 cm^{-1} at $\vec{k}=0$, the Davydov-split Γ_1^+, Γ_2^+ excitons at 169 cm^{-1} and 204 cm^{-1}, and the $\Gamma_3^+ + \Gamma_4^+$ branch at 194 cm^{-1}. In addition there are four (Γ_1^+, Γ_2^+), and four ($\Gamma_3^+ + \Gamma_4^+$) branches around 800 cm^{-1} which have not yet been observed by any technique. Light scattering is allowed by symmetry from all