ANISOTROPIC COUPLING BETWEEN MAGNETIC IONS IN TERBIUM

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Abstract

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The magnon energies in the c-direction of Tb have been studied as a function of magnetic field applied in the easy and hard directions in the basal plane. From the results, the components of the anisotropic two-ion coupling between the moments have been deduced as a function of $\vec{q}$. There is a very large anisotropy between the hexagonal axis and the basal plane which persists as the temperature is increased, and a smaller anisotropy in the plane which decreases with increasing temperature. It is tentatively suggested that the anisotropy in the two-ion coupling is primarily magneto-elastic in origin.

1. INTRODUCTION

The study of the magnons in rare earth metals by inelastic neutron scattering has proved to be a rich source of information on the magnetic interactions in these materials [1]. The early studies on Tb were principally concerned with the exchange interactions between the magnetic ions, and the mechanisms through which these contributed to the stability of the different magnetic phases [2]. It was tacitly assumed that the magnetic anisotropy was due to the crystal fields in the unstrained lattice and could be written in a simple single-ion form. Later experiments on the temperature dependence [3], and particularly the magnetic field dependence [4], of the magnon energies showed, however, that this assumption is not strictly valid. Measurements on a Tb-10% Ho single crystal [4] showed unambiguously that magnetoelastic effects, due to the distortion of the lattice on magnetization, contribute to the single-ion anisotropy and actually dominate the hexagonal component. Furthermore, it was shown that the lattice strain is unable to follow the rapid precession of the moment when a magnon is excited, so that the 'frozen-lattice' model is valid. These experiments have recently been repeated for pure Tb [5] with similar results.

In the interpretations of most of the spin-wave measurements which have so far been reported on the rare earths it was further assumed that the exchange can be represented by a simple isotropic Heisenberg Hamiltonian. However, in a recent study of the conical magnetic phase of Er, Nicklow et al. [6] demonstrated that their results could only be explained quantitatively if the usual spin-wave Hamiltonian were augmented by a large anisotropic two-ion coupling. This intriguing conclusion prompted us to extend our measurements of the field dependence of the magnon energies to finite q-values, and thereby to study the spatial dependence of the anisotropic coupling between magnetic ions in Tb.

In the next section, we discuss the Hamiltonian which we have used to describe our results, and from it derive theoretical expressions for the magnon energies in an external magnetic field. These expressions are used in section 3 to interpret the experimental results and to derive from them values for the anisotropic two-ion coupling. Finally we make a few comments on the physical significance of our conclusions.

2. THE HAMILTONIAN AND MAGNON DISPERSION RELATIONS

The various contributions to the spin-wave Hamiltonian comprise an exchange coupling between the moments, which we allow to be anisotropic, a single-ion anisotropy term $\mathcal{H}_s$, including magnetoelastic effects, which we do not need to specify in detail, and a two-ion magnetoelastic coupling. In addition, we include a Zeeman term due to an external magnetic field. The total Hamiltonian then takes the form

$$\mathcal{H} = \mathcal{H}_x + \mathcal{H}_s + \mathcal{H}^{II}_{me} - g\beta \sum_i \underline{J}_i \cdot \underline{H} \qquad (1)$$

We use a set of Cartesian axes such that ζ is along the hexagonal axis and ξ is the easy direction of magnetization in the plane. The terms representing the coupling between the magnetic ions may then be written

$$\mathcal{H}_x = - \sum_{i>j} \left\{ \mathcal{J}^{aa}_{ij} J_{i\eta} J_{j\eta} + \mathcal{J}^{bb}_{ij} J_{i\xi} J_{j\xi} + \mathcal{J}^{cc}_{ij} J_{i\zeta} J_{j\zeta} \right\} \qquad (2)$$

$$\mathcal{H}^{II}_{me} = - \sum_{i>j} \left\{ 3^{\frac{1}{2}} D^{\alpha}_{ij} \bar{\varepsilon}^{\alpha} \left[J_{i\zeta} J_{j\zeta} - \tfrac{1}{3} \underline{J}_i \cdot \underline{J}_j \right] \right.$$
$$\left. - D^{\gamma}_{ij} \left[(J_{i\xi} J_{j\xi} - J_{i\eta} J_{j\eta}) \bar{\varepsilon}^{\gamma}_1 + (J_{i\xi} J_{j\eta} + J_{i\eta} J_{j\xi}) \bar{\varepsilon}^{\gamma}_2 \right] \right\} \qquad (3)$$

In these expressions the notation generally follows that of ref. [1]. The D_{ij} are two-ion magnetoelastic coupling coefficients [7] and the $\bar{\varepsilon}$ are equilibrium strains, implying that we are using the now well-established frozen-lattice model. For simplicity, we have considered only the lowest-order strains in the magnetoelastic terms.

When the magnetic field is applied in the easy direction, the moments remain aligned in this direction and the Hamiltonian (1) may be diagonalized by standard techniques to give the magnon energies

$$\varepsilon^2_H(\underline{q}) = (A_o + B_o + A_{\underline{q}} + B_{\underline{q}} + g\beta H)(A_o + A_{\underline{q}} - B_o - B_{\underline{q}} + g\beta H) \qquad (4)$$

A_o and B_o involve only the single-ion anisotropy terms, while $A_{\underline{q}}$ and $B_{\underline{q}}$ are defined by

$$A_{\underline{q}} + B_{\underline{q}} = J \left[K^{cc}(o) - K^{cc}(\underline{q}) \right]$$
$$A_{\underline{q}} - B_{\underline{q}} = J \left[K^{aa}(o) - K^{aa}(\underline{q}) \right] \qquad (5)$$

where

$$K^{cc}(\underline{q}) = \sum_j \left\{ \mathcal{J}_{ij}^{cc} + 2\cdot 3^{-3/2}(2D+G)D_{ij}^{\alpha} \right\} e^{i\underline{q}\cdot(\underline{R}_i-\underline{R}_j)}$$

$$K^{aa}(\underline{q}) = \sum_j \left\{ \mathcal{J}_{ij}^{aa} + 3^{-3/2}(2D+G)D_{ij}^{\alpha} + CD_{ij}^{\gamma} \right\} e^{i\underline{q}\cdot(\underline{R}_i-\underline{R}_j)} \quad (6)$$

C, D and G are magnetostriction coefficients [8]. Since we are interested principally in the dispersion relations in the c-direction, we will use the double-zone representation [1] for the hcp structure throughout.

When the field is applied in the hard direction the moments turn towards the field and, at a critical value H_c, they become aligned with the field. Above this field, the magnon dispersion relations are also given by an expression of the form (4), except that H is replaced by $(H-H_c)$ and a is replaced by b in (5) and (6). We note however that higher-order two-ion magnetoelastic effects can give rise to a difference between $K^{bb}(\underline{q})$ and $K^{aa}(\underline{q})$.

3. EXPERIMENTAL RESULTS AND INTERPRETATION

The magnon energies in Tb were measured as a function of applied magnetic field at three different temperatures by inelastic neutron scattering at the DR 3 reactor at Risø. The sample was a monocrystalline disc with a diameter of about 14 mm and a thickness of about 5 mm, with the c-axis normal to the faces. An external field of up to 48 kG could be applied by means of a superconducting solenoid. A selection of the results for different $\underline{q}$-values in the c-direction is shown in Fig. 1.

From these results, we may readily deduce the functions $K(\underline{q})$ which describe the anisotropic coupling between the magnetic ions. For a field in the easy direction, we find by differentiating (4) that, in the limit of small H

$$\alpha_{\underline{q}} = \frac{d\,\varepsilon^2(\underline{q})}{dH} = 2g\beta(A_o + A_{\underline{q}}) \quad (7)$$

If the two-ion anisotropy is negligible, so that $K^{cc}(\underline{q})$ and $K^{bb}(\underline{q})$ are identical, then from (5) $B_{\underline{q}}$ vanishes so that, using (4)

$$\alpha_{\underline{q}}/g\beta = 2\,[\varepsilon_o^2(\underline{q}) + B_o^2]^{1/2} \quad (8)$$

The two sides of this equation, calculated from the experimental results, are compared in Fig. 2, and from their difference we can immediately deduce that the two-ion anisotropy is finite. Including $B_{\underline{q}}$, we obtain from (4)

$$B_o + B_{\underline{q}} = \pm\left[\left(\frac{\alpha_{\underline{q}}}{2g\beta}\right)^2 - \varepsilon_o^2(\underline{q})\right]^{1/2} \quad (9)$$

and from (7) and (9) we find

$$J\ [K^{cc}(o) - K^{cc}(\underline{q})] = A_{\underline{q}} + B_{\underline{q}}$$

$$= \pm \left[\left(\frac{a_{\underline{q}}}{2g\beta} \right)^2 - \varepsilon_o^2(\underline{q}) \right]^{1/2} + \frac{a_{\underline{q}}}{2g\beta} - (A_o + B_o) \qquad (10)$$

$$J\ [K^{aa}(o) - K^{aa}(\underline{q})] = A_{\underline{q}} - B_{\underline{q}}$$

$$= \frac{a_{\underline{q}}}{2g\beta} \mp \left[\left(\frac{a_{\underline{q}}}{2g\beta} \right)^2 - \varepsilon_o^2(\underline{q}) \right]^{1/2} - (A_o - B_o) \qquad (11)$$

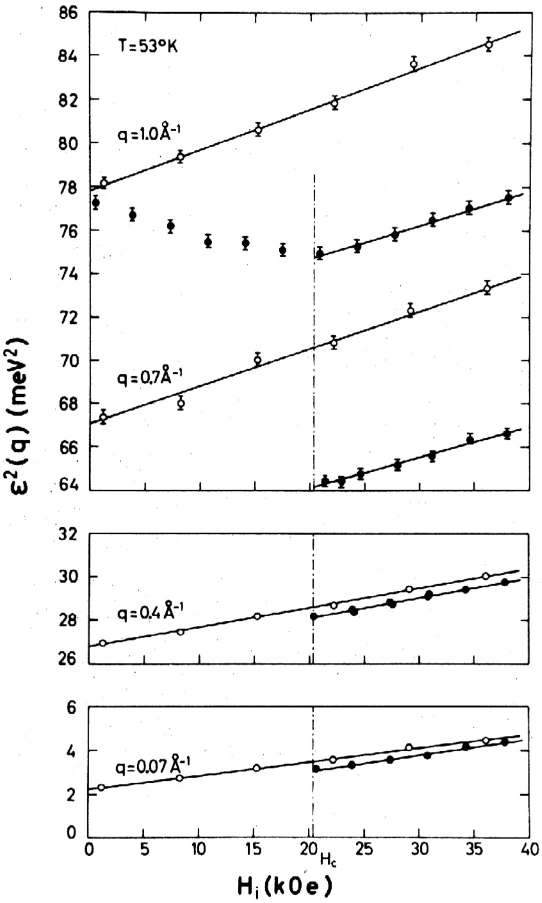

FIG. 1. The dependence of the square of the magnon energies for different $\vec{q}$ values in the c-direction on internal magnetic field in Tb at 53°K. Open symbols represent results for the field in the easy direction, and closed for the field in the hard direction.

The ambiguity in signs in (9)-(11) can lead to difficulties of interpretation when the expression within the square root becomes zero. We have taken the signs which seem most consistent with our experimental results. The alternative choice of signs leads to results which are qualitatively similar but quantitatively different from those shown in Fig. 3. This point is being investigated further by means of more experimental measurements.

We note that the single-ion anisotropy enters only through the parameters A_o and B_o, which are independent of $\underline{q}$. A similar analysis for a field in the hard direction allows the determination of $K^{cc}(\underline{q})$ and $K^{bb}(\underline{q})$.

In Fig. 3 we have plotted the values of the two-ion coupling functions deduced from our experimental results at 53°K and 134°K. There is a large anisotropy between the c-axis and the basal plane which persists as the temperature is increased, and a smaller anisotropy in the plane which decreases significantly with increasing temperature. The results for $K^{cc}(\underline{q})$ deduced from measurements with the field in the easy and hard directions are satisfactorily consistent. At 134°K there is a pronounced anomaly in the planar $K(\underline{q})$, in the form of a very rapid increase near the zone-boundary.

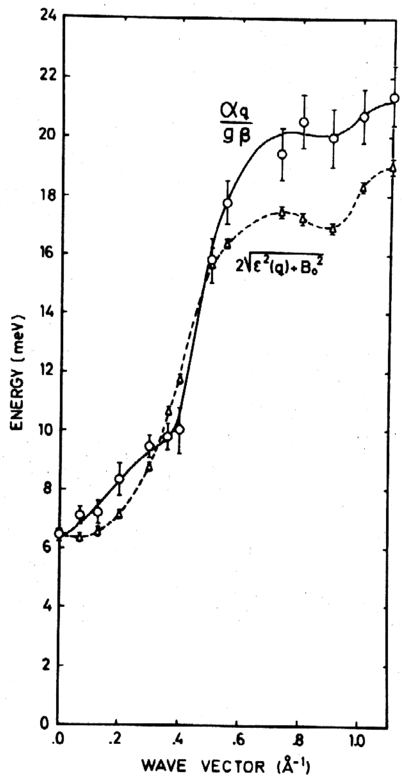

FIG. 2. A comparison between the functions $\alpha_{\vec{q}}/g\beta$ and $2[\epsilon_0^2(\vec{q})+B_0^2]^{\frac{1}{2}}$ for the c-direction of Tb at 4.2°K. The difference between them reveals the presence of anisotropy in the two-ion coupling.

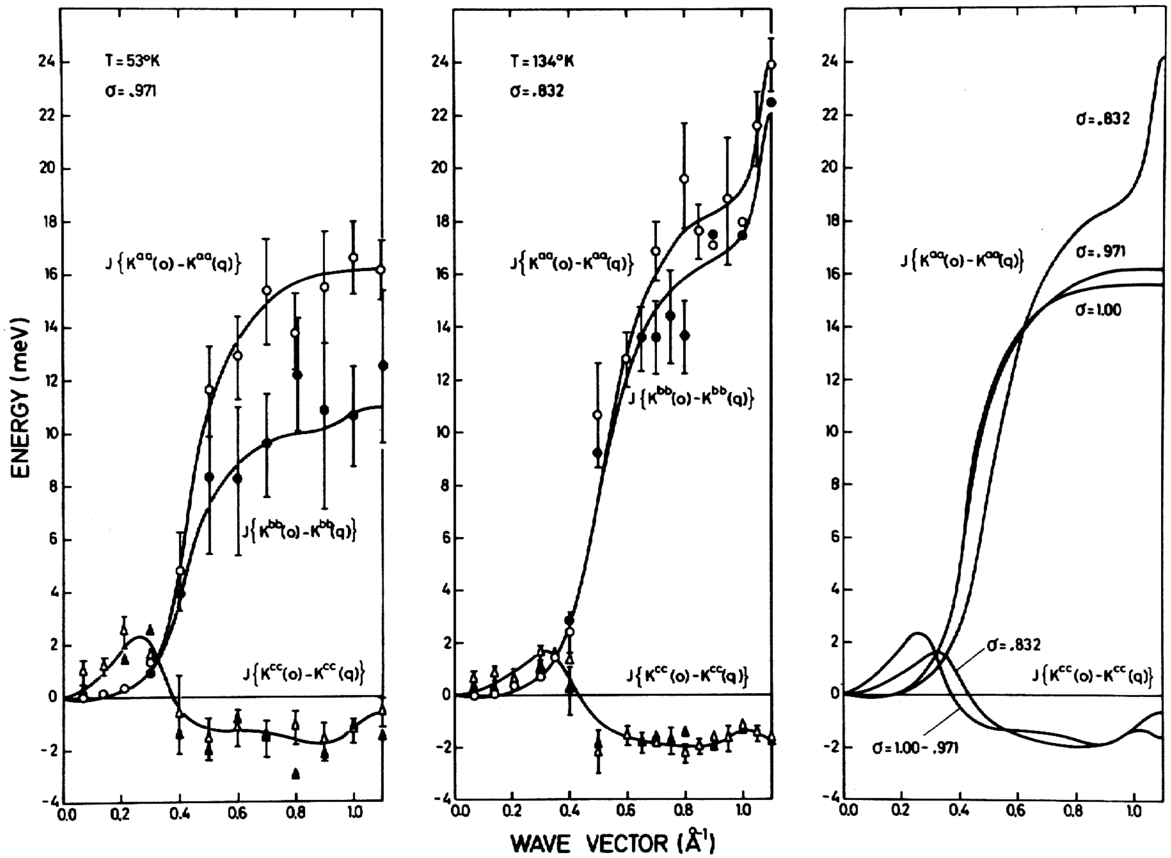

FIG. 3. The Fourier transforms of the effective two-ion coupling between moments for the c-direction of Tb at different temperatures. Open symbols are derived from measurements with the field in the easy direction, and closed for the field in the hard direction.

4. DISCUSSION

The most striking feature of these results is the very large difference between $K^{cc}(q)$ and the planar components. Anisotropic exchange can result from spin-orbit coupling or polarization of the conduction electron gas, but these would both tend to single out the magnetization direction, which is also the direction of the orbital moment. It seems likely therefore that the observed anisotropy is due to the magnetoelastic coupling between the lattice and the magnetic system. However the two-ion magnetoelastic coupling terms in (6) should decrease with relative magnetization [9] at least as rapidly as σ^4, and the persistence of the axial anisotropy as the temperature is increased is therefore puzzling. On the other hand the magnetoelastic contributions to the planar anisotropy should decrease at least as rapidly as σ^8, and this is consistent with our observations. The development of an anomaly near the zone boundary in the planar components may be associated with the change in electronic structure with magnetic polarization. The extension of these measurements to a wider temperature range and the study of different rare earth systems should provide an improved understanding of the pronounced anisotropy in the coupling between the magnetic ions.

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DISCUSSION

R.A. COWLEY: Were spin-wave interactions included in your analysis and, if not, how would their inclusion affect your results?

H. BJERRUM MØLLER: Spin-wave interactions are not included in our analysis. The renormalization effects should be reflected in the temperature dependence of the resulting $K(q)_s$: there should be a decrease with increasing temperature, in contrast to the result obtained.