# Anisotropic exchange interaction in the conical magnetic phase of erbium

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Abstract. From a general two ion spin Hamiltonian, we deduce an expression for the energies of spin waves propagating in a hexagonal solid in which the magnetic moments are ordered in a conical or helical structure. The spin wave dispersion relation in the c direction of Er in its conical magnetic phase at 4.5 K, which has been studied by Nicklow et al (1971) is reanalysed. In this analysis we introduce an alternative kind of anisotropic coupling between the total angular moments  $(J_i \text{ and } J_i)$  on the sites i and j, proportional to the following combination of Racah operators:  $\tilde{O}_{2,-2}(J_i)$ ,  $\tilde{O}_{2,-2}(J_j)$ , expressed with respect to a coordinate system with the z axis along the c direction. The resulting anisotropy (both the constant and the q dependent part) is reduced by an order of magnitude in comparison with that deduced by Nicklow et al (1971). The constant anisotropy is found to be equal to about 20 meV (a rough estimate from magnetization measurements gives 5–10 meV). The anisotropic part of the exchange interaction is found to be of the same order of magnitude as the isotropic part.

### 1. Introduction

The energies,  $\epsilon(q)$ , for spin waves propagating in the c direction of Er at 4.5 K have been measured by Nicklow et al (1971) (cited as I hereafter). At this temperature, the ionic moments of Er are ordered in a conical configuration which is specified by a wave vector,  $\mathbf{Q}$ , and a cone angle  $\theta$ . Neutron diffraction studies (Cable et al 1965) have revealed that  $\mathbf{Q}$  is parallel to the c axis and equal to  $0.24 \times 2\pi/c$ , and  $\theta$ , the angle between the magnetic moment on each site and the c direction, is equal to  $28.5^{\circ}$  for Er at 4.5 K.

The magnetic excitations in Er are spin waves. On account of the conical ordering the basal plane is no longer a mirror plane, which implies that the energies in general will be different for spin waves propagating parallel, +q, and antiparallel, -q, to the ferromagnetic component of the magnetic moments, ie  $\epsilon(+q) \neq \epsilon(-q)$ . The cone angle in Er is quite small, which allows the splitting between  $\epsilon(+q)$  and  $\epsilon(-q)$  to be clearly observed as in the results of I.

In I the spin wave measurements are interpreted by means of the model Hamiltonian generally used for describing the magnetic properties of the heavy rare earth metals (Cooper et al 1962, Cooper 1968, Goodings and Southern 1971). In this model the splitting between the two spin wave branches gives a rather direct determination of the isotropic part of the exchange interaction,  $\mathcal{J}(q)$ . The spin wave energies can then be satisfactorily reproduced only by including a very large anisotropic exchange in the form of an effective axial two ion coupling (Cooper et al 1962). This result contrasts

with the results obtained later for the basal plane ferromagnets Tb (Jensen and Houmann 1973) and Dy (Nicklow and Nielsen, private communication), in which the isotropic and anisotropic components of the exchange interaction are of the same order of magnitude.

The apparent importance of the anisotropic exchange interaction in the heavy rare earth metals requires a more precise and systematic treatment of the influence of two ion anisotropy on the energy spectrum of the spin waves. In the next section we shall consider the case of conically (and helically) ordered spin system.

## 2. Anisotropic exchange interaction

The ionic moments in the rare earth metals are coupled indirectly through the conduction electrons. In the simple RKKY model, this coupling takes the form of an isotropic Heisenberg interaction:

$$\mathcal{H}_{H} = -\frac{1}{2} \sum_{i \neq j} \mathcal{J}(\mathbf{R}_{i} - \mathbf{R}_{j}) \mathbf{J}_{i} \cdot \mathbf{J}_{j}$$
 (1)

where  $J_i$  is the total angular moment on site  $R_i$ . Various orbital effects (the spin-orbit coupling of the conduction electrons, electric multipole interactions, and the influence of the ionic orbital moment on the s-f exchange matrix element) may contribute to the effective Hamiltonian for the spin system and give rise to anisotropy (Wolf 1971, Levy 1969, Elliott and Thorpe 1968). Without discussing the microscopic origins we can write a general spin Hamiltonian phenomenologically as

$$\mathcal{H}_{IJ} = \frac{1}{2} \sum_{i \neq j} \sum_{l, m} \sum_{l' = m'} K_{lm'}^{l'm'} (\mathbf{R}_i - \mathbf{R}_j)$$

$$\times \left[ \tilde{O}_{l, m}(J_i) \tilde{O}_{l' = m'}(J_i) + (-1)^{m+m'} \tilde{O}_{l' = m}(J_i) \tilde{O}_{l' = m'}(J_i) \right] \qquad m \geqslant 0$$
(2)

where  $\tilde{O}_{l,m}(J_i)$  are spherical tensor (Racah) operators (Wolf 1971. Danielsen and Lindgård 1972, Buckmaster et al 1972). A more general Hamiltonian should include terms where the coefficients depend on the spin polarization at site  $R_i$  (Levy 1969, Jensen et al). The number of independent terms is limited by the general restrictions (Wolf 1971): l and l' smaller than or equal to seven for f electrons, and l+l' even. For spin waves propagating in the c direction of a hexagonal solid (in the case of an hcp structure we can use the double zone representation), the effective spin Hamiltonian is reduced by the symmetry requirements, m+m' equal to 0 or  $\pm$  6 (or  $\pm$  12), when referring to a coordinate system with the c axis along the c direction. The introduction of these conditions in (2) results in 102 independent parameters,  $K_{lm}^{l'm'}$  (including the four single ion terms). We shall here only consider the two ion terms for which the ranks of both tensors (l and l') are smaller than three. These terms, which presumably are the most important in (2), will reflect the behaviour of the terms of higher rank.

The notation is simplified by defining the Fourier transforms as

$$K_{lm}(\boldsymbol{q}) = \sum_{i} K_{lm}^{l-m}(\boldsymbol{R}_{i} - \boldsymbol{R}_{j}) \exp\left[i\boldsymbol{q} \cdot (\boldsymbol{R}_{i} - \boldsymbol{R}_{j})\right]$$
(3)

Formally the single ion Hamiltonian is included in (2) ( $\tilde{O}_{0,0} = 1$ ) by defining

$$V_{lm} = \sum_{i} K_{lm}^{00}(\boldsymbol{R}_{i} - \boldsymbol{R}_{j}). \tag{4}$$

The spin Hamiltonian (2) is now treated by a conventional spin wave technique (Cooper et al 1962, Cooper 1968, Goodings and Southern 1971). The Racah operators are trans-

formed (Danielsen and Lindgård 1972, Buckmaster et al 1972) into a coordinate system in which the ordered moment (of the cone) is always along the local z axis. The Racah operators are then expanded in spin deviation operators (Danielsen and Lindgård 1972), and only those terms which are either independent of or quadratic in the spin deviation operators are retained. If the planar anisotropy,  $V_{66}$ , is neglected, the Hamiltonian is diagonalized by a Fourier transformation followed by an appropriate canonical transformation of the spin deviation operators to magnon operators ( $\alpha_a$ )

$$\mathcal{H}_{JJ} = (NJ)E + \sum_{q} \epsilon(q)\alpha_{q}^{+}\alpha_{q}$$
 (5)

where the reduced equilibrium energy (N is the number of ions in the crystal) is given by

$$E = \frac{1}{2}J_{1}V_{20}(3\cos^{2}\theta - 1) + \frac{1}{8}J_{3}V_{40}(35\cos^{4}\theta - 30\cos^{2}\theta + 3) + \frac{1}{16}J_{5}V_{60}(231\cos^{6}\theta - 315\cos^{4}\theta + 105\cos^{2}\theta - 5) + JK_{10}(0)\cos^{2}\theta - \frac{1}{2}JK_{11}(\mathbf{Q})\sin^{2}\theta + \frac{1}{4}JJ_{1}^{2}K_{20}(0) \times (3\cos^{2}\theta - 1)^{2} - \frac{8}{8}JJ_{1}^{2}K_{21}(\mathbf{Q})\sin^{2}2\theta + \frac{8}{8}JJ_{1}^{2}K_{22}(2\mathbf{Q})\sin^{4}\theta$$
(6)

where we have used the abbreviation

$$J_n = (J - \frac{1}{2})(J - 1)\dots(J - \frac{1}{2}n). \tag{7}$$

The stability of the cone structure requires the partial derivatives of E with respect to Q and  $\theta$  to be zero. Using the last condition we can write the energy of spin waves propagating in the c direction as follows:

$$\epsilon(\mathbf{q}) = C(\mathbf{q}) + [F_1(\mathbf{q}) F_2(\mathbf{q})]^{1/2}$$
(8)

where the first term, C(q), changes sign when q is changed to -q

$$C(\mathbf{q}) = \cos \theta \left\{ \frac{1}{2} J \left[ K_{11} (\mathbf{Q} + \mathbf{q}) - K_{11} (\mathbf{Q} - \mathbf{q}) \right] + \frac{3}{2} J J_{1}^{2} \left[ K_{21} (\mathbf{Q} + \mathbf{q}) - K_{21} (\mathbf{Q} - \mathbf{q}) \right] \cos 2\theta - \frac{3}{2} J J_{1}^{2} \left[ K_{22} (2\mathbf{Q} + \mathbf{q}) - K_{22} (2\mathbf{Q} - \mathbf{q}) \right] \sin^{2} \theta \right\}$$

$$(9)$$

which then is equal to half the splitting between the two magnon branches. Both C(q) and  $F_1(q)$  go to zero when q goes to zero, and they are both independent of terms in (2) in which m is equal to zero:

$$F_{1}(\mathbf{q}) = \frac{1}{2}J[2K_{11}(\mathbf{Q}) - K_{11}(\mathbf{Q} + \mathbf{q}) - K_{11}(\mathbf{Q} - \mathbf{q})] + \frac{3}{2}JJ_{1}^{2}[2K_{21}(\mathbf{Q}) - K_{21}(\mathbf{Q} + \mathbf{q}) - K_{21}(\mathbf{Q} - \mathbf{q})]\cos^{2}\theta - \frac{3}{2}JJ_{1}^{2}[2K_{22}(2\mathbf{Q}) - K_{22}(2\mathbf{Q} + \mathbf{q}) - K_{22}(2\mathbf{Q} - \mathbf{q})]\sin^{2}\theta$$
(10)

and finally

$$F_{2}(\mathbf{q}) = \frac{1}{2}J[2K_{11}(\mathbf{Q}) - K_{11}(\mathbf{Q} + \mathbf{q}) - K_{11}(\mathbf{Q} - \mathbf{q})]\cos^{2}\theta + \frac{3}{2}JJ_{1}^{2}[2K_{21}(\mathbf{Q}) - K_{21}(\mathbf{Q} + \mathbf{q}) - K_{21}(\mathbf{Q} - \mathbf{q})]\cos^{2}2\theta - \frac{3}{2}JJ_{1}^{2}[2K_{22}(2\mathbf{Q}) - K_{22}(2\mathbf{Q} + \mathbf{q}) - K_{22}(2\mathbf{Q} - \mathbf{q})] \times \sin^{2}\theta \cos^{2}\theta + \{-2J[K_{10}(0) - K_{10}(\mathbf{q})] - 18JJ_{1}^{2}[K_{20}(0) - K_{20}(\mathbf{q})]\cos^{2}\theta + L\}\sin^{2}\theta$$
(11)

where L is a q independent constant composed of single and two ion contributions

$$L = 3J_1V_{20} + \frac{15}{2}J_3V_{40}(7\cos^2\theta - 1) + \frac{105}{8}J_5V_{60}(33\cos^4\theta - 18\cos^2\theta + 1) + 2JK_{10}(0) + JK_{11}(\mathbf{Q}) + 3JJ_1^2K_{20}(0) (9\cos^2\theta - 1) + 3JJ_1^2K_{21}(\mathbf{Q}) (6\cos^2\theta - 1) + \frac{3}{2}JJ_1^2K_{22}(2\mathbf{Q})(3\cos^2\theta - 1).$$
 (12)

Equations (5)–(12) also account for the case of a helically ordered system as the special case of a cone structure where  $\theta = \pi/2$  (in this particular case C(q) vanishes identically).

In I the spin wave dispersion relation was analysed using an expression for the energies which corresponds to (5)–(12) when  $K_{21}(q)$  and  $K_{22}(q)$  are neglected. However, the behaviour of the  $K_{21}$  and  $K_{22}$  terms differs essentially from the behaviour of the  $K_{11}$  term. The  $K_{21}$  term differs by the  $\theta$  dependence (notice that  $F_2(q)$  still depends on  $K_{21}(Q \pm q)$  for a helix), and the  $K_{22}$  term by the wave vector argument,  $2Q \pm q$ . These differences imply that a simple relation between C(q) and C(q) does not exist when C(q) or C(q) are introduced in the spin Hamiltonian

In general all wave vector arguments,  $m\mathbf{Q} \pm \mathbf{q}$ , may occur in all three magnon energy parameters (9)–(11) arising from the  $K_{lm}^{l'-m}$  term in (2).  $K_{l0}^{l'0}$  will be present only in  $F_2(\mathbf{q})$ . In the particular case of a helical structure we have a simple selection rule:  $K_{lm}^{l'-m}$  will contribute to  $F_1(\mathbf{q})$  when l+m is even and  $m \neq 0$ , and to  $F_2(\mathbf{q})$  when l+m is odd.

The terms  $K_{lm}^{l'6-m}$  can (in this context) be taken into account by introducing a q dependent hexagonal anisotropy,  $J_5V_{66}(q)$ . The effect of this term has been considered in some detail by Cooper et al (1962). When  $J_5V_{66}(q)$  is weak in comparison with  $F_1(q)$  and  $F_2(q)$ , this term will give rise to a mixing of the magnon modes with wave vector q and  $q \pm 6Q$ . This mixing may result in energy gaps at crossing points of the unperturbed dispersion relations,  $\epsilon(q)$  and  $\epsilon(q \pm 6Q)$ .

# 3. Spin waves in Er

We have analysed the spin wave measurements in I on the basis of the expressions (6)–(12) deduced above. The isotropic exchange interaction (1) is introduced in (6)–(12) by putting

 $K_{11}(q) = \mathcal{J}(q)$   $K_{10}(q) = -\frac{1}{2}\mathcal{J}(q)$  (13)

As shown in I, it is not possible to obtain any reasonable fit to the experimental dispersion relation without introducing two ion anisotropy. The spin wave energies can be reproduced equally well with any one of the three possibilities:  $K_{2m}(q)$  identical to zero unless m is equal to 0, 1, or 2 (the ambiguity of the fit if more than one anisotropy parameter is introduced leaves us with those three alternatives). However, the use of the information obtained from the neutron intensities observed at a given constant q scan (figure 1 in I) combined with magnetization measurements (Rhyne et al 1968) enables us to conclude that  $K_{2,2}(q)$  is the most important two ion anisotropy term in Er.

Without knowing the crystal field parameters,  $V_{l0}$ , we can estimate the magnitude of L (and  $V_{60}$ ) from the critical fields necessary to pull the spins into ferromagnetic alignment parallel with the field when the field is applied along the c direction,  $H_{\parallel,c}$ , and in the basal plane,  $H_{\perp,c}$  (assuming the transitions to be of second order):

$$L = \sin^2\theta \left\{ 2g\mu_{\mathbf{B}}(H_{\perp,c} + H_{\parallel,c}\cot^4\theta) - 2J[K_{11}(\mathbf{Q}) - K_{11}(0)] + 6JJ_1^2[K_{21}(\mathbf{Q}) - K_{21}(0)] + 3JJ_1^2[K_{22}(2\mathbf{Q}) - K_{22}(0)] \right\}.$$
(14)

The two critical fields (Rhyne et al 1968) are both of the order of 200 kOe and, using the final values for the exchange parameters (which in (14) contribute with -0.8 meV) we obtain L equal to about 7 meV. In the same approximation we find that  $J_5V_{60}$  is equal to about 0.08 meV, which is in fair agreement with the value of 0.05 meV deduced by Chikazumi et al (1971) from torque measurements on a dilute alloy of Er in Gd. When a field is applied in the basal plane, the cone collapses into a fan structure at a field of 17 kOe: when the field is further increased to  $H_{\perp,f} \simeq 45$  kOe, the fan phase is converted into a ferromagnetic state with the moments lying at an angle near that of the original cone (Rhyne et al 1968). This implies that

$$K = J[K_{11}(\mathbf{Q}) - K_{11}(0)] + 3JJ_1^2[K_{21}(\mathbf{Q}) - K_{21}(0)]\cos 2\theta$$
$$-\frac{3}{2}JJ_1^2[K_{22}(2\mathbf{Q}) - K_{22}(0)]\sin^2\theta = \frac{g\mu_B H_{11}}{\sin\theta}$$
(15)

which gives a value  $K \simeq 0.63$  meV. The effects of magnetoelastic interaction (in general the effects of terms in the spin hamiltonian (2) where  $K_{lm}^{l'm'}$  depends on the spin polarization) have been neglected in (14)–(15) so that the values obtained for L and K are only representative.

The analysis in I, in which  $K_{21}(q)$  and  $K_{22}(q)$  are neglected, leads to values of L and K (L=104 meV and K=0.08 meV) which are wrong by an order of magnitude compared with those deduced from magnetization measurements. Furthermore, the theoretical ratio between the neutron intensities (Baryakhtar and Maleev 1963) of the magnon branches +q and -q does not behave in agreement with the observed intensities (for q larger than Q) as stated in I.

In the present work most of these discrepancies have been removed by introducing  $K_{22}(q)$  instead of  $K_{20}(q)$ . The exchange couplings were expressed in terms of cosine series with interplanar exchange constants as coefficients:

$$J[\mathcal{J}(0) - \mathcal{J}(\mathbf{q})] = \sum_{n} \mathcal{J}_{n}[1 - \cos(\zeta n\pi)]$$
(16)

and we define an effective anisotropy by

$$J[\mathcal{K}_{22}(0) - \mathcal{K}_{22}(\mathbf{q})] = -3JJ_1^2[K_{22}(0) - K_{22}(\mathbf{q})] = \sum_{n} \mathcal{K}_n[1 - \cos(\zeta n\pi)]$$
 (17)

where  $\zeta = qc/2\pi$ . These exchange constants and L were used as variables in a least squares fitting to the measured dispersion relation. The final fit and the experimental magnon energies are shown in figure 1. The results of the interplanar exchange constants,  $\mathscr{J}_n$  and  $\mathscr{K}_n$ , and L are given in table 1 together with their uncertainties. In figure 2 is shown the q dependence of the isotropic exchange interaction,  $J\mathscr{J}(q)$ , and of the two ion anisotropy,  $J\mathscr{K}_{22}(q)$ . The values obtained for L and K (L=20 meV and K=0.98 meV) are comparable with the values deduced from (14) and (15). The exchange energy, (6), has its minimum at q=Q as should be the case. The behaviour of the calculated intensities is in agreement with the neutron groups shown in figure 1 in I except in the neighbourhood of q=Q. We calculate a 4.5.1 intensity ratio for the magnons at q=+2Q and -2Q: at larger q this ratio increases to 7.5.1. The experimentally observed intensity ratios are 4:1 and approx. 10:1 respectively.

The neutron intensities obtained experimentally at q = +Q and -Q suggest a ratio equal to  $1\cdot 2:1$  (whereas we calculate a ratio of 5:1). This discrepancy indicates that some other couplings are present, otherwise  $F_2(Q)$  should be 300 times larger than  $F_1(Q)$ , which seems very unlikely. This disagreement between the theoretical and the

experimental intensity ratio may be due to several mechanisms. Antisymmetric exchange interaction (Levy 1969) may modify the simple relation between the intensities and  $F_1(q)$  and  $F_2(q)$ . The magnon (at a wave vector q) may no longer be a pure mode owing to the interaction with the phonons (Nayyar and Sherrington 1972) at wave vectors  $q \pm nQ$  and with the magnons at wave vectors  $q \pm nQ$  where n is equal to 1, 2, ...

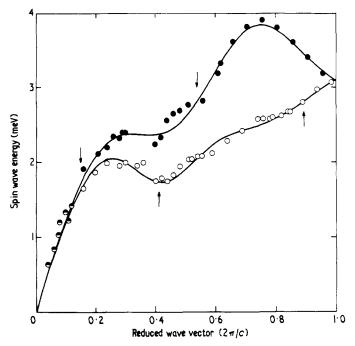


Figure 1. Spin wave energies  $\epsilon(+q)$  and  $\epsilon(-q)$  (open and closed symbols respectively) in the c direction of Er at 4.5 K (after Nicklow et al 1971). The solid lines are the result of the least squares fit to equations (8)–(11) in the text. The arrows mark the points where  $\epsilon(q) = \epsilon(q \pm 6Q)$ .

Using the point charge value for the hexagonal anisotropy  $(J_5V_{66} = -0.08 \text{ meV})$  we obtain energy gaps equal to about 0.05 meV in the magnon spectrum at points where  $\epsilon(q) = \epsilon(q \pm 6Q)$  (these points are marked by arrows in figure 1). The amplitude of this interaction may be even larger, and some of the irregularities of the measured magnon energies may be due to this coupling. Because  $\epsilon(q + 6Q) \simeq \epsilon(-q)$  when q is

Table 1	. The interplanar	exchange con	nstants and I	All values are	in meV
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Isotropy constants	Anisotropy constants	
$J_1 + 0.901 \pm 0.16$	$\mathcal{K}_1 - 0.423 \pm 0.42$	
$f$ , $-0.265 \pm 0.02$	$\mathcal{K}_2 - 0.408 \pm 0.16$	
$I_3 - 0.036 \pm 0.04$	$\mathcal{K}_3 - 0.476 \pm 0.14$	
$I_4 - 0.167 \pm 0.20$	$\mathcal{K}_4 + 0.204 \pm 0.68$	
$I_5 - 0.162 \pm 0.06$	$\mathcal{K}_{5} - 0.616 \pm 0.18$	
$I_6 - 0.038 \pm 0.02$	$L + 20.0 \pm 6.0$	

close to Q, this interaction will enhance the apparent intensity of the -q branch at the expense of the intensity of the +q branch and may thus be part of the explanation of the abnormal intensity ratio observed at q = Q.

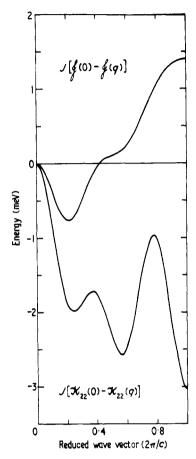


Figure 2. The q dependence of the isotropic and the anisotropic components of the exchange interaction in the c direction of Er.

The constant L could be constrained to be equal to 7 meV with the consequences of a slight worsening of the fit to the dispersion relation, of an unreasonably large K value, and of a poor agreement with the observed intensity ratios. Nor if  $K_{20}(q)$  and  $K_{22}(q)$  are both set equal to zero is a satisfactory fit obtained, because the minimum in the exchange energy occurs at q = Q/2 instead of at q = Q, and because of a drastic change of the isotropic part of the exchange interaction, which takes a form quite different from that observed in the other heavy rare earths.

#### 4. Conclusion

The replacement of the effective axial two ion coupling,  $K_{20}(q)$ , by the two ion anisotropy,  $K_{22}(q)$ , yields a substantial improvement in the description of the properties of the

spins in Er. The present analysis does not exclude the existence of other kinds of anisotropic exchange interactions; however, we conclude that  $K_{2,2}(\mathbf{q})$  is the most important coupling. A more definite conclusion can be drawn only on the basis of additional information about the spin system, eg the spin wave energies as function of field applied along the symmetry directions. The q dependence of the isotropic part of the exchange interaction,  $\mathcal{J}(q)$ , is close to that obtained in I, except that the maximum near the wave vector of the conical structure is much more pronounced. Apart from this maximum which is essential for stabilizing the periodic magnetic structure in Er, the appropriately scaled function,  $[\mathcal{J}(0) - \mathcal{J}(q)]/(g-1)^2$ , is very similar, both in magnitude and q dependence, to that deduced for Gd (Koehler et al 1970), where the exchange presumably only has an isotropic component. The exchange anisotropy is reduced by a factor of 20 compared with the result in I; however, it is still rather substantial.  $J\mathcal{K}_{22}(q)$  agrees both in sign and in absolute magnitude with the axial anisotropy deduced in Tb (Jensen and Houmann 1973), which indicates some systematic behaviour of the anisotropic exchange couplings in the heavy rare earth metals analogous to the scaling of the isotropic exchange energies with respect to the de Gennes factor  $(q-1)^2 J(J+1)$ , which still appears as an adequate scaling.

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