



**HAL**  
open science

# FIELD DEPENDENCE OF THE SPIN WAVE ENERGY IN RARE-EARTH METALS

P. Lindgård

► **To cite this version:**

P. Lindgård. FIELD DEPENDENCE OF THE SPIN WAVE ENERGY IN RARE-EARTH METALS. Journal de Physique Colloques, 1971, 32 (C1), pp.C1-238-C1-240. 10.1051/jphyscol:1971177 . jpa-00214504

**HAL Id: jpa-00214504**

**<https://hal.science/jpa-00214504>**

Submitted on 4 Feb 2008

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

## FIELD DEPENDENCE OF THE SPIN WAVE ENERGY IN RARE-EARTH METALS

P. A. LINDGÅRD

A. E. C., Risø, Roskilde, Denmark

**Résumé.** — L'approximation du réseau rigide, discutée par B. R. Cooper, est élaborée. Une expression de la relation de dispersion des ondes de spin est faite au moyen des coefficients d'une expansion complète en harmoniques sphériques du potentiel local dans le réseau (h. c.) distordu. Un calcul des charges de point est effectué pour le terbium utilisant les déformations expérimentales. Les mesures des ondes de spin dans Tb-10 % Ho par M. Nielsen et al. sont analysées.

**Abstract.** — The « frozen-lattice » approximation, discussed by Cooper has, been extended. The spin wave energy is expressed in terms of the coefficients of a complete expansion in spherical harmonics of the local potential in the distorted h c p. lattice. These coefficients are related to the macroscopic first and second order magnetostriction constants. A point charge calculation has been performed for terbium, using the values for the lattice distortion obtained from magnetostriction measurements. Spin wave energy gap measurements by M. Nielsen et al. on a Tb-10 % Ho crystal are analyzed. Second order magneto-elastic effects are of importance.

**Introduction.** — The spinwaves in the rare earth metals depend strongly on the magnetic anisotropy which is partly of electrostatic and partly of magneto-elastic origin. The magnetoelastic contribution depends on the orientation of the magnetization and may be varied by a magnetic field. The anisotropy in the rare earth has been discussed previously [1] for an ideal h. c. p. lattice and the magnetostriction was considered by Tsuya *et al.* [2]. In the following the effect of a magnetic field on the spinwave spectrum will be investigated. It is necessary to include higher order magneto-elastic contribution in order to explain the recent spinwave measurements on terbium by Nielsen *et al.* [3].

**The Crystal Field.** — The « frozen lattice » approximation was proposed by Turov and Shavrov [4] and discussed at length by Cooper [5]. In this approximation it is supposed that the lattice is uniformly strained under the influence of the average magnetic moment, but that the lattice is rigid under the high frequent spin precession in a spinwave. The spinwaves therefore depend on the deformed crystal field. For small strains the potential at an electron at position  $\mathbf{r}$  is

$$V(\mathbf{r}) = V_0(\mathbf{r}) + \sum_i \bar{\epsilon}_i \partial_i \partial \epsilon_i V_0(\mathbf{r}) = V_0(\mathbf{r}) + \delta V(\mathbf{r}),$$

where  $V_0(\mathbf{r})$  is the unperturbed potential and  $\bar{\epsilon}_i$  are the strain components in the lattice corresponding to the equilibrium distortions.  $\delta V(\mathbf{r})$  can be expanded in spherical harmonics using the knowledge of the unperturbed symmetry. The strain  $\epsilon_1^y = 1/2(\epsilon_{\xi\xi} - \epsilon_{\eta\eta})$  is the strain caused by a magnetic moment in the (1, 0, 0, 0) or the (1, 2, 0, 0) direction in the hexagonal basal plane.  $\xi$  is the easy direction of magnetization ; the b-direction (1, 2, 0, 0) for terbium and the a-direction (1, 0, 0, 0) for dysprosium. The potential due to the displaced ionic point charges is for this strain :

$$\delta V(\mathbf{r}) = -Ze^2 \sum_{l,m}' (-1)^m Y_l^{-m}(\hat{\mathbf{r}}) \times \left\{ \sum_n K_l(R_n, r) [Y_2^2(\hat{\mathbf{R}}_n) + Y_2^{-2}(\hat{\mathbf{R}}_n)] Y_l^m(\hat{\mathbf{R}}_n) \right\} \bar{\epsilon}_1^y \quad (1)$$

where  $Z$  is the ionic charge and  $K_l(R_n, r)$  involves summations over 3  $j$  symbols and the ratios of  $r$  and  $R_n$ . The sum over the  $n$  unperturbed neighbour positions  $R_n$  gives only contributions for the allowed values of  $L, M$  for the unperturbed symmetry. The possible values of  $l$  and  $m$  for  $\delta V(\mathbf{r})$  to which the  $\Sigma'$  is restricted are, from (1),  $l = L + 2$  ;  $m = M \pm 2$  with  $L = 0, 2, 4, 6$  and  $M = 0, \pm 6$  for the hexagonal lattice. At a 4 f electron in the rare earth metal there will also be a contribution to the electric field from the conduction electrons. To a first approximation this may be assumed to possess the same symmetry causing a uniform screening, replacing  $Z$  by a  $Z_{\text{eff}}$ .

**Phenomenological Hamiltonian.** — The distorted potential  $V_0(\mathbf{r}) + \delta V(\mathbf{r})$  can on the basis of the symmetry (1) in the usual way [6] be expressed in terms of spinoperators and the resulting spinhamiltonian for the basal plane strains  $\epsilon_1^y$  and  $\epsilon_2^y = \epsilon_{\xi\eta}$  is then  $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{me}}$  :

$$\mathcal{H}_0 = -\sum_{i,j} J_{ij} \mathbf{S}_i \otimes \mathbf{S}_j + \sum_i \left\{ \sum_{L=2,4,6} B_L^0 \mathcal{O}_{Li}^0 + B_6^6 \mathcal{O}_{6i}^6 - g\mu(H_{\perp} S_{\eta i} + H_{\parallel} S_{\xi i}) \right\}, \quad (2)$$

$$\mathcal{H}_{\text{me}} = \sum_i \sum_{l,m}' B_l^m \{ \mathcal{O}_l^m(c)_i \bar{\epsilon}_1^y + \mathcal{O}_l^m(s)_i \bar{\epsilon}_2^y \}, \quad (3)$$

where  $l = 2, 4, 6$  and  $m = 2, 4$ , and  $\mathcal{O}_l^m(c, s)$  are the spinoperators, some of which are listed by Hutchings [6] and a few exemplified below.  $B_l^m$  are phenomenological constants. The point charge contribution to these will be a geometrical factor times  $\langle r^l \rangle \theta_l$ , the 4 f radial distribution integrals and the Stevens factors [6].

The classical phenomenological hamiltonian of Callen and Callen [7] including the  $l = 2$  and 4 terms is

$$\mathcal{H}_{\text{me}}^c = -\tilde{B}^{\gamma,2} \sum_i \{ (\mathbf{S}_{\xi i}^2 - \mathbf{S}_{\eta i}^2) \bar{\epsilon}_1^y + 2 \mathbf{S}_{\xi i} \mathbf{S}_{\eta i} \bar{\epsilon}_2^y \} - \tilde{B}^{\gamma,4} \sum_i \{ (\mathbf{S}_{\xi i}^4 - 6 \mathbf{S}_{\xi i}^2 \mathbf{S}_{\eta i}^2 + \mathbf{S}_{\eta i}^4) \bar{\epsilon}_1^y - 4 \mathbf{S}_{\xi i} \mathbf{S}_{\eta i} (\mathbf{S}_{\xi i}^2 - \mathbf{S}_{\eta i}^2) \bar{\epsilon}_2^y \}.$$

It involves only the spinfunctions  $\mathcal{O}_2^2(c, s)$  and  $\mathcal{O}_4^4(c, s)$  of eq (3). The directional dependence of the magnetoelastic energy  $H_{me}$  can be measured and gives the following relations between the parameters in (3) and (4) :

$$\begin{aligned} \mathcal{S}_2 B_2^2 - \mathcal{S}_4 B_4^2 + \mathcal{S}_6 B_6^2 &= - \mathcal{S}_2 \tilde{B}^{\gamma,2} = - cC, \\ \mathcal{S}_4 B_4^4 - \mathcal{S}_6 B_6^4 &= - \mathcal{S}_4 \tilde{B}^{\gamma,4} = - \frac{1}{2} cA, \end{aligned} \quad (5)$$

where  $c = c^2/S$  is the reduced elastic constant,  $A, C$ , are the magnetostriction constants defined and measured by Rhyne and Legvold [8] and

$$\mathcal{S}_l = \langle \mathcal{O}_l^0 \rangle = \left( S - \frac{1}{2} \right) \times (S - 1) \dots \left( S - \frac{l-1}{2} \right).$$

**Spinwaves in a Magnetic Field.** — The spinwave dispersion relation is obtained by a diagonalization [9] of the complete hamiltonian (2) and (3) and the expression is reduced using the relations (5).

If a field  $H_{\perp}$  is applied in the hard direction (1, 0, 0, 0) in terbium it rotates the moment to an angle  $\varphi$  from this. By an energy minimum condition on (2) and (3) and using (5),  $\varphi$  is determined by

$$g\mu H_{\perp} = 12 \cos \varphi (4 \cos^2 \varphi - 1) (4 \cos^2 \varphi - 3) \times (-B_6^c \mathcal{S}_6 + \frac{1}{2} cAC). \quad (6)$$

The corresponding spinwave energy is given by

$$\begin{aligned} (\hbar\omega_q)_{\varphi}^2 &= \{ E_q - 2 P_2 S + c(2 C^2 + A^2) - \Delta M \cos 6 \varphi + \\ &+ \frac{1}{6} g\mu H_c (1 + 2 \cos 2 \varphi + 2 \cos 4 \varphi) \} \times \\ &\times \{ E_q + 4 c(C^2 + A^2 + 2 AC \cos 6 \varphi) + \\ &+ \frac{1}{6} g\mu H_c (1 + 2 \cos 2 \varphi + \\ &+ 2 \cos 4 \varphi - 5 \cos 6 \varphi) \}, \end{aligned} \quad (7)$$

where  $E_q = 2S(J_0 + J'_0 - J_q \pm |J'_q|)$  is the exchange energy and  $P_2$  the effective axial anisotropy, used by Cooper.  $P_2$  involves terms with  $m = 0$  and  $l = 2, 4, 6$  and also a contribution from the higher order magnetoelastic terms in (3).

The field  $H_c$  necessary to pull the moment into the hard direction is obtained from (6) for  $\varphi = 0$ . The last term in (6) is of magnetoelastic origin and is dominant in terbium [3]. For fields larger than  $H_c$ ,  $\varphi = 0$  and the spinwave energy is given by (7) after an addition of  $g\mu(H_{\perp} - H_c)$  in both braces.

For a magnetic field  $H_{\parallel}$  in the easy direction,  $\varphi = \pi/2$  and the spinwave energy is obtained from (7) after the addition of  $g\mu H_{\parallel}$  in both braces. The magnetoelastic term  $\Delta M$  is the contribution from the higher order terms in (3) and gives the only qualitative difference in the spinwave expression between the complete Hamiltonian (3) and the classical (4). For large axial anisotropy, therefore, the classical Hamiltonian (4) is a very good approximation as it gives the correct parallel molecular field (last brace in (7)) and only an error in the first brace, which is dominated by the axial contribution. However,  $\Delta M$  gives rise to a different  $\varphi$ -dependence of (7). If  $\Delta M$  and  $A$  are neglected, (7) reduce to the formula obtained by Cooper. The spinwave measurements on terbium [3] show that it is essential to include the second order magnetoelastic constant  $A$ . Very good agreement is then obtained with the macroscopic magnetostriction constants  $A$  and  $C$  [8]. The  $\Delta M$  term could not be determined for terbium which was to be expected because of the large axial anisotropy. A similar situation is likely to occur for dysprosium. The application of a moderate magnetic field in the basal plane of holmium changes it to a b-axis ferromagnet [10]. In holmium the axial anisotropy is not dominant and the higher order terms contributing to  $\Delta M$  may be of importance. From (7) it is seen that the sign of  $AC$  is important in determining the relative magnitudes of  $(\hbar\omega_0)_{easy}$  and  $(\hbar\omega_0)_{hard}$ .

**Point Charge Calculation.** — The contribution to  $B_l^m$  in (2) and (3) from a set of nearest neighbour effective point charges, in the strained Tb lattice, has been computed numerically, using the actual ion positions. These can be deduced from measurements of the lattice constants [12] and the magnetostriction [8].

Table I shows the experimental [3] and theoretical values at  $T = 0$ . For simplicity the full charge  $Z = 3$  and the free ion  $\langle r^l \rangle$  values, calculated in the Hartree Föch approximation [12] have been used. The experimental values decrease much faster with increasing order and  $l$  than the theoretical prediction. The  $l = 6$  term was zero within the experimental accuracy. This shows that an expansion of the 4f cloud in the crystal, as conjectured by Kasuya [1] is not the important mechanism. A possible explanation of the difference between the calculations and the experimental result is that the conduction electrons are not uniformly distributed and contribute to the potential in which

TABLE I

*Comparison between the experimental results and a point charge calculation for terbium.  $Z = 3$  and the Freeman-Watson [12] values for  $\langle r^l \rangle$  have been used. The experimental parameters decrease faster with increasing  $l$  than predicted by the point charge model. The difference between the easy- and hard direction data is due to effects nonlinear in strain. The units are meV/atom.*

Order of $l$	$- 2 P_2 S$	$cC$ , eq. (5)	$cA$ , eq. (5)	$- 36 B_6^c \mathcal{S}_6$
—	2	2	4	6
—	—	—	—	—
Experiment ref. [3]	4.8	12.3	4.6	< 0.01
Theory, easy direction	2.4	4.8	3.4	0.12
Theory, hard direction	2.6	3.1	3.5	0.13

the 4 f electrons move. Spectroscopic data and band calculations for the rare earth metals show that the lowest lying conduction electron state has the 5 d character. Occupation of orbitals of  $(x \pm iy)^2$  symmetry would increase the screening in the plane, increase the axial anisotropy and simultaneously reduce the sixfold anisotropy. A strain dependent occupation of xz and yz orbitals will increase the contribution to the  $m = 2$  term, that is the contribution to  $C$  (eq. 5). With a screening of this kind there is room for an expansion of the wavefunctions counter-weighted by a reduction of  $Z$ .

**Summary.** — The spinwave dispersion relation for a strained rare earth metal has been expressed in terms of the parameters of an expansion of the perturbed potential in spherical harmonics (eq. 3).

This expansion is complete as far as effects on the 4 f electrons are concerned when terms up to  $l = 6$  are included. The parameters are related to macroscopic magnetostriction constants (eq. 5) and are also calculated in a point charge model for terbium. The inclusion of the second order magnetoelastic constant is essential in order to explain the measurement of the spinwave energy gap in terbium [3]. The point charge calculation gives the correct sign and order of magnitude for the  $l = 2$  and 4 anisotropy parameters in terbium. Comparison with experimental results gives no evidence of a significant expansion of the 4 f electron cloud as has been suggested by Kasuya [1]. An effect which will possibly improve the numerical values is an anisotropic screening due to a non-uniform strain dependent conduction electron density.

#### References

- [1] KASUYA (T.), *Magnetism* vol II. B, ed. Rado and Suhl, Academic Press, 1966.
- [2] TSUYA (N.), CLARK (A. E.) and BOZORTH (R. M.), 250, *Proc. Int. Conf. on Magnetism*, Nottingham, sept. 1964.
- [3] NIELSEN (M.), MØLLER (H. B.), LINDGÅRD (P. A.) and MACKINTOSH (A. R.) *Phys. Rev. Letters*, 1970, **25**, 1451.
- [4] TUROV (E. A.) and SHAROV (V. G.), *Sovjet Phys. Solid State*, 1966, **7**, 166.
- [5] COOPER (B. R.), *Phys. Rev.*, 1968, **169**, 281.
- [6] See for example HUTCHINGS (M. T.), *Solid State Phys.*, 1964, **16**, 227.
- [7] CALLEN (E.) and CALLEN (H. B.), *Phys. Rev.*, 1965, **139**, A 455.
- [8] RHYNE (J. J.) and LEGVOLD (A.), *Phys. Rev.*, 1965, **138**, A 507.
- [9] See for example LINDGÅRD (P. A.), KOWALSKA (A.) and LAUT (P.), *Phys. Chem. Sol.*, 1967, **28**, 1357.
- [10] KOEHLER (W. C.), CABLE (J. W.), CHILD (H. R.), WILKINSON (M. K.) and WOLLAN (W. O.), *Phys. Rev.*, 1967, **158**, 450.
- [11] DARNELL (F. J.), *Phys. Rev.*, 1963, **132**, 128.
- [12] FREEMAN (A. J.) and WATSON (R. E.), *Phys. Rev.*, 1962, **127**, 2058.