J. Phys. C: Solid State Phys., Vol. 8, 1975. Printed in Great Britain.

Hartree–Fock renormalization of magnetic anisotropy in the basal-plane ferromagnets terbium and dysprosium

Jens Jensen
Department of Theoretical Physics, 12 Parks Road, Oxford OX1 3PQ

Received 24 March 1975

Abstract. We show that the application of the Holstein–Primakoff transformation in the spin-wave theory of an anisotropic ferromagnet implies contributions from the isotropic two-ion coupling to the energy of the uniform spin-wave mode. Including these contributions, the energies of the HF renormalized spin waves are deduced using the method of Lindgård and Danielsen. The macroscopic resonance theory for the uniform mode is found to agree with the spin-wave theory if the magnetization is along a symmetry direction. Numerical results for the renormalization of the static anisotropy parameters of Tb and Dy are presented. The effect of the ellipticity of the moment precession is found to be smaller than reported previously.

1. Introduction

The temperature dependence of the magnetic anisotropy parameters in the ferromagnetic phases of the two rare-earth metals Tb and Dy has been the subject of a recent paper by Lindgård and Danielsen (1975), to be referred to as I. They used a generalized Holstein–Primakoff transformation (Lindgård and Danielsen 1974) to obtain an expansion of the magnetic Hamiltonian in terms of spin deviation operators. The expansion was performed such that it accounts systematically for kinematic effects (well ordered expansion). A self-consistent Hartree–Fock (HF) decoupling was then utilized in a calculation of the renormalization of single-ion Stevens operators and the energy of the uniform spin-wave excitation.

The renormalization of the macroscopic anisotropy parameters was found to second order in $\Delta M(T)$, the magnetization–deviation parameter, and $b(T)$, which characterizes the non-sphericity of the moment precession. Their results represent an extension of the work of Brooks and co-workers (Brooks *et al* 1968, Brooks and Egami 1973) who also found deviations from the classical $l(l+1)/2$ power law to first order in $b(T)$.

In this paper we shall follow closely the approach of Lindgård and Danielsen (1975). However, it is shown that the two-ion coupling introduces an adjustment of the magnon energy gap at zero wavevector, removing the unphysical features of the Holstein–Primakoff transformation reported by Brooks (1970) and present in the calculation in *I*. Within the HF approximation, we find that the energy gap is determined strictly by the differential change of the free energy, which may be deduced from torque and magnetostriction measurements. This is in agreement with that deduced by Brooks (1970) using an alternative spin-operator expansion of the magnetic Hamiltonian. Here we discuss briefly the dynamic corrections to this result which seems to be of some importance in Tb (Houmann *et al* 1975).

Finally, we include a discussion of quantitative results obtained for Tb and Dy. These results are based on the correlation functions, $\Delta M(T)$ and $b(T)$, calculated in *I*. The temperature dependences of the anisotropy parameters are found to be closer to the $l(l+1)/2$ power law than reported previously. A calculation of the energy gap in Dy from static anisotropy parameters agrees with the experimental value, supporting the proposal of Egami (1972) of the importance of zero-point effects on the hexagonal anisotropy in Dy.

2. Spin-wave theory in the Hartree–Fock approximation

The two heavy rare-earth metals, Tb and Dy (HCP structure), are both ferromagnetically ordered at low temperatures. The axial anisotropy in the two metals is very large (Rhyne and Clark 1967, Ferron *et al* 1970) and the ionic angular moments are strongly confined to the basal plane. The non-cylindrical symmetry of the magnetic potential affects the normal modes and gives rise to deviations of the ground state from a fully aligned spin state at zero temperature.

The magnetic Hamiltonian in these metals is known to be very complex (Jensen *et al* 1975). For simplicity we shall neglect the presence of two-ion anisotropy and thus consider the two-ion coupling to be isotropic

$$\mathscr{H}_{\mathrm{II}} = -\sum_{i,j} \mathscr{J}(i,j)\boldsymbol{J}_i \cdot \boldsymbol{J}_j, \tag{1}$$

where $\boldsymbol{J}_i$ is the total angular moment on site i. The presence of two ions per unit cell is inessential in this context and we shall consider the Fourier-transformed two-ion coupling to be described by one parameter, $\mathscr{J}(\boldsymbol{q})$, only.

The magnetic anisotropy, including magnetoelastic anisotropy, is then due to the single-ion Hamiltonian

$$\mathscr{H}_{\mathrm{I}} = \mathscr{H}_{\mathrm{CF}} + \mathscr{H}_{\mathrm{Z}}, \tag{2}$$

where $\mathscr{H}_{\mathrm{Z}}$ is the Zeeman term

$$\mathscr{H}_{\mathrm{Z}} = -g\mu_{\mathrm{B}} \sum_i \boldsymbol{J}_i \cdot \boldsymbol{H} \tag{3}$$

and $\mathscr{H}_{\mathrm{CF}}$ is the crystal-field Hamiltonian. We shall follow the notation used in *I*, and express $\mathscr{H}_{\mathrm{CF}}$ in terms of Stevens operators:

$$\mathscr{H}_{\mathrm{CF}} = \sum_i \left[\sum_{l=2,4,6} B_l^0 O_l^0(c) + B_6^6 O_6^6(c) \right]_i + \mathscr{H}_{\mathrm{me}}. \tag{4a}$$

The expression for $\mathscr{H}_{\mathrm{me}}$ (the magnetoelastic anisotropy) can be found in I (equation 4). The Stevens operators appearing in (4*a*) are defined with respect to the c axis as the polar axis which also defines the polar angles θ and φ. Rotating the quantization axis so as to be along the direction of magnetization, $\mathscr{H}_{\mathrm{CF}}$ may be written

$$\mathscr{H}_{\mathrm{CF}} = \sum_i \left[\sum_{l,m} K_l^m(\theta,\varphi) O_l^m(c) \right]_i, \qquad l \text{ and } m \text{ even}, \tag{4b}$$

(see *I*) where the coefficients K_l^m depend on the polar angles.

The total magnetic Hamiltonian, $\mathscr{H} = \mathscr{H}_{\mathrm{I}} + \mathscr{H}_{\mathrm{II}}$, is transformed into a well ordered power expansion in spin deviation operators by applying the method of the generalized

Holstein–Primakoff transformation (equation (26) of I) derived by Lindgård and Danielsen (1974):

$$\mathscr{H}_{\mathrm{I}} = E_{\mathrm{I}} + \sum_i [Aa_i^+ a_i + B\tfrac{1}{2}(a_i^+ a_i^+ + a_i a_i) + C_1 a_i^+ a_i^+ a_i a_i + C_2(a_i^+ a_i^+ a_i^+ a_i + a_i^+ a_i a_i a_i) + C_3(a_i^+ a_i^+ a_i^+ a_i^+ + a_i a_i a_i a_i)], \tag{5a}$$

where only terms to fourth order in the spin deviation operator are retained. The coefficients in (5*a*) may all be expressed in terms of K_l^m defined by (4*b*). A similar expansion of $\mathscr{H}_{\mathrm{II}}$ gives

$$\mathscr{H}_{\mathrm{II}} = E_{\mathrm{II}} + \sum_{\boldsymbol{q}} 2J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{q})]a_{\boldsymbol{q}}^+ a_{\boldsymbol{q}} + N^{-1} \sum_{\boldsymbol{q}\boldsymbol{q}_1\boldsymbol{q}_2} \tfrac{1}{2}\mathscr{J}(\boldsymbol{q})[a_{\boldsymbol{q}}^+ a_{\boldsymbol{q}}^+ a_{\boldsymbol{q}_2} a_{\boldsymbol{q}+\boldsymbol{q}_1-\boldsymbol{q}_2} + a_{\boldsymbol{q}_1} a_{\boldsymbol{q}_2}^+ a_{\boldsymbol{q}} a_{\boldsymbol{q}_1+\boldsymbol{q}_2-\boldsymbol{q}} - 2a_{\boldsymbol{q}_1}^+ a_{\boldsymbol{q}_2}^+ a_{\boldsymbol{q}_1+\boldsymbol{q}} a_{\boldsymbol{q}_2-\boldsymbol{q}}]. \tag{5b}$$

In order to calculate the thermal expectation value of the Stevens operators, $\langle O_l^m(c)\rangle$, the total Hamiltonian is decoupled by means of a Hartree–Fock approximation which is the analogue of a random-phase decoupling of the equations of motion. The spin deviation operators obey the boson relations by which the following equation is deduced:

$$[a_{\boldsymbol{q}}, \mathscr{H}] = \{A + 2J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{q})]\}a_{\boldsymbol{q}} + Ba_{-\boldsymbol{q}}^+ + N^{-1} \sum_{\boldsymbol{k},\boldsymbol{k}'} \{[-2\mathscr{J}(\boldsymbol{q}-\boldsymbol{k}') + \mathscr{J}(\boldsymbol{k}') + \tfrac{1}{2}\mathscr{J}(\boldsymbol{k}) + \tfrac{1}{2}\mathscr{J}(\boldsymbol{q}) + 2C_1]a_{\boldsymbol{k}}^+ a_{\boldsymbol{k}'} a_{\boldsymbol{q}+\boldsymbol{k}-\boldsymbol{k}'} + C_2(3a_{\boldsymbol{k}}^+ a_{-\boldsymbol{k}'}^+ a_{\boldsymbol{q}+\boldsymbol{k}-\boldsymbol{k}'} + a_{-\boldsymbol{k}} a_{\boldsymbol{k}'} a_{\boldsymbol{q}+\boldsymbol{k}-\boldsymbol{k}'}) + 4C_3 a_{\boldsymbol{k}}^+ a_{-\boldsymbol{k}'}^+ a_{-\boldsymbol{q}-\boldsymbol{k}+\boldsymbol{k}'}^+\}. \tag{6}$$

Defining

$$\Delta M(T) = N^{-1} \sum_{\boldsymbol{k}}' m_{\boldsymbol{k}}, \qquad m_{\boldsymbol{k}} = J^{-1}\langle a_{\boldsymbol{k}}^+ a_{\boldsymbol{k}}\rangle, \tag{7a}$$

$$b(T) = N^{-1} \sum_{\boldsymbol{k}} b_{\boldsymbol{k}}, \qquad b_{\boldsymbol{k}} = J^{-1}\langle a_{\boldsymbol{k}}^+ a_{-\boldsymbol{k}}^+\rangle = J^{-1}\langle a_{\boldsymbol{k}} a_{-\boldsymbol{k}}\rangle, \tag{7b}$$

then equation (6) is reduced by the random-phase approximation to

$$[a_{\boldsymbol{q}}, \mathscr{H}] = A_{\boldsymbol{q}}(T)a_{\boldsymbol{q}} + B_{\boldsymbol{q}}(T)a_{-\boldsymbol{q}}^+, \tag{8a}$$

where

$$A_{\boldsymbol{q}}(T) = A + 4JC_1\Delta M + 6JC_2 b + 2J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{q})] \times (1 - \Delta M) + N^{-1} \sum_{\boldsymbol{k}} 2J[\mathscr{J}(\boldsymbol{k}) - \mathscr{J}(\boldsymbol{k}-\boldsymbol{q})]m_{\boldsymbol{k}} \tag{8b}$$

and

$$B_{\boldsymbol{q}}(T) = B + 2JC_1 b + 6JC_2\Delta M + 12JC_3 b + N^{-1} \sum_{\boldsymbol{k}} J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{k})]b_{\boldsymbol{k}} - J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{q})]b + N^{-1} \sum_{\boldsymbol{k}} 2J[\mathscr{J}(\boldsymbol{k}) - \mathscr{J}(\boldsymbol{k}-\boldsymbol{q})]b_{\boldsymbol{k}}. \tag{8c}$$

The use of the well ordered power expansion in spin deviation operators ensures that the approximation introduced by (5*a*) is well defined, as the terms neglected contribute to (8) only in higher power of ΔM and b. By the random-phase approximation, the Hamiltonian is transformed into one for which the elementary excitations are non-interacting renormalized spin waves, as (8*a*) is diagonalized by a Bogoliubov transformation

$$\mathscr{H} = E_0 + \sum_{\boldsymbol{q}} E_{\boldsymbol{q}}(T)\alpha_{\boldsymbol{q}}^+ \alpha_{\boldsymbol{q}} \tag{9a}$$

where the renormalized excitation energies are determined by

$$E_q(T) = \{[A_q(T) + B_q(T)][A_q(T) - B_q(T)]\}^{1/2}. \tag{9b}$$

This enables us to express the characteristic correlation functions (equation 7) self-consistently in terms of the final temperature-dependent energy parameters:

$$m_k = \frac{1}{J}\left\{\frac{A_k(T)}{E_k(T)}[n_k(T) + \tfrac{1}{2}] - \tfrac{1}{2}\right\}, \tag{10a}$$

$$b_k = -\frac{1}{J}\frac{B_k(T)}{E_k(T)}[n_k(T) + \tfrac{1}{2}], \tag{10b}$$

as given in I (equation 32). $n_k(T)$ is the boson population factor.

At zero wavevector, all the two-ion contributions to $A_0(T)$ and $B_0(T)$ vanish except for one term in (8*c*):

$$\Delta B_0^{II}(T) = N^{-1}\sum_k J[\mathscr{J}(0) - \mathscr{J}(k)]b_k. \tag{11}$$

Using equations (8) and (10), $\Delta B_0^{II}(T)$ can be written

$$\Delta B_0^{II}(T) = -\tfrac{1}{4}J^{-1}B - \tfrac{1}{2}B\Delta M - \tfrac{1}{2}Ab. \tag{12}$$

Including this contribution, the temperature dependence of the energy gap at zero wavevector is changed in comparison with the expression given in I (equations (33) and (34)). Using the parameters

$$\alpha_l = l(l+1)/2 \text{ and } \qquad S_l = J(J - \tfrac{1}{2})\ldots(J - \tfrac{1}{2}(l-1)) \tag{13}$$

defined in I and $C(l, m)$ given in table 2 in I, we find to first order in J^{-1}, ΔM and b

$$A_0(T) \pm B_0(T) = J^{-1}\sum_l S_l\{-K_l^0 C(l,0)\alpha_l[1 - (\alpha_l - 1)\Delta M \mp \tfrac{1}{2}\alpha_l b]$$
$$\pm K_l^2 C(l,2)[1 - (\alpha_l - 1)\Delta M \mp (\alpha_l - \tfrac{3}{2})b] \pm K_l^4 C(l,4)12b\}. \tag{14}$$

Notice that $\Delta M/J$ and b/J have to be considered as terms of second order because of the neglection of higher-order two-ion contributions in (11). In the derivation of (14), no first-order terms are neglected, as is done in equation (34) of I, in an attempt to obtain agreement with the Goldstone theorem. In the next section we show that the expression (14) for the energy gap fulfills this theorem, eg $A_0(T) + B_0(T)$ vanishes identically in the case of pure planar anisotropy.

The introduction of the two-ion contribution (11) to the spin-wave energy gap at zero wavevector has removed the unphysical features of the Holstein–Primakoff transformation present in the calculation in I, and which are also discussed by Brooks (1970). Brooks avoided this difficulty by studying instead the equations of motion of J^+ and J^- using a Wortis expansion of J_z. Because $\Delta B_0^{II}(T)$ appears in (8*c*) as a term independent of q, it should be interpreted as a single-ion term, and a comparison with the work of Brooks shows that the occurrence of this term is a characteristic of the Holstein–Primakoff transformation.

3. Relationship between static and dynamic energy gap parameters

In I, the thermal expectation value of a Stevens operator was evaluated by taking the trace over the non-interacting spin-wave states, (9*a*). The details can be found in I; we shall only quote the result (equation 36):

$$\begin{aligned}
\langle O_l^0(c)\rangle_T &= S_l C(l,0)[1 - \alpha_l \Delta M + \tfrac{1}{2}\alpha_l(\alpha_l - 1)(1 + \tfrac{1}{2}J^{-1})(\Delta M^2 + \tfrac{1}{2}b^2)] \\
\langle O_l^2(c)\rangle_T &= S_l C(l,2)(1 + \tfrac{1}{4}J^{-1})b[1 - (\alpha_l - \tfrac{3}{2} - \tfrac{3}{8}J^{-1})(1 + \tfrac{1}{2}J^{-1})\Delta M] \\
\langle O_l^4(c)\rangle_T &= S_l C(l,4)(1 + \tfrac{3}{2}J^{-1})6b^2
\end{aligned} \tag{15}$$

where the Stevens operators are those defined with respect to the direction of magnetization as the polar axis, (4*b*). J^{-1} is an independent expansion parameter, and the expressions are evaluated to first order in J^{-1} and to second order in ΔM and b.

The magnetic anisotropy determined by torque or magnetization measurements is related to the change of the free energy, $F(\theta, \varphi)$, as a function of the polar angles. In the case where the magnetic excitations can be considered to be non-interacting bosons (equation 9*a*), the free energy is reduced to

$$F(T) = E_0 + k_B T \sum_q \ln [1 - \exp(-E_q/k_B T)]. \tag{16}$$

As also shown by Goodings and Southern (1970), this implies that the first derivative of $F(T)$ at constant temperature with respect to φ for example is

$$\left.\frac{\partial F}{\partial \varphi}\right|_\theta = \left.\frac{\partial U}{\partial \varphi}\right|_\theta - \sum_q E_q \left.\frac{\partial n_q}{\partial \varphi}\right|_\theta . \tag{17}$$

The internal energy, $U = \langle \mathscr{H}_I + \mathscr{H}_{II} \rangle$, is a function of (θ, φ) and also of m_k and b_k, which depend implicitly on the angles. Substituting (15) into (4*b*) and calculating $\langle \mathscr{H}_{II} \rangle$ within the same approximation, the following relations are easily derived:

$$\left.\frac{\partial U}{\partial m_k}\right|_{\theta,\varphi;b_k} = JA_k(T), \qquad \left.\frac{\partial U}{\partial b_k}\right|_{\theta,\varphi;m_k} = JB_k(T). \tag{18}$$

The introduction of these relations in (17) leads to the important equation

$$\left.\frac{\partial F}{\partial \varphi}\right|_\theta = \left.\frac{\partial U}{\partial \varphi}\right|_{\theta;m_k,b_k} \tag{19}$$

which allows us to calculate the derivatives of the free energy from a knowledge of $U(\theta, \varphi)$. When the magnetization is along a symmetry direction, we have immediately from (19)

$$\begin{aligned}
F_{\varphi\varphi} \equiv \left.\frac{\partial^2 F}{\partial \varphi^2}\right|_\theta &= \left.\frac{\partial^2 U}{\partial \varphi^2}\right|_{\theta;m_k,b_k}, & \varphi &= p\frac{\pi}{6} \\
F_{\theta\theta} \equiv \left.\frac{\partial^2 F}{\partial \theta^2}\right|_\varphi &= \left.\frac{\partial^2 U}{\partial \theta^2}\right|_{\varphi;m_k,b_k}, & \theta &= p\frac{\pi}{2}
\end{aligned} \tag{20}$$

because in these cases all the first derivatives with respect to the angles vanish identically (and $F_{\theta\varphi} = 0$).

A phenomenological macroscopic theory of ferromagnetic resonance has been

developed by Smith and Beljers (1955) in which the energy of the uniform spin-wave mode is found to be

$$E_{q=0} = g\mu_B M^{-1}[F_{\theta'\theta'} F_{\varphi'\varphi'} - F^2_{\theta'\varphi'}]^{1/2}, \tag{21}$$

where θ' and φ' are angles defined with respect to a polar axis perpendicular to the magnetization $\boldsymbol{M}$:

$$M = Ng\mu_B J\sigma \qquad \sigma = 1 - \Delta M. \tag{22}$$

If magnetoelastic couplings are present, the second derivatives have to be evaluated at constant strains (Jensen 1971), introducing a slight complication in the use of (21), as a knowledge of the magnetostriction coefficients is required besides the information obtained from magnetization measurements (constant stress conditions).

The validity of (21) within the spin-wave approximation is easily shown in the case of an axial ferromagnet (Brooks and Egami 1973) as $B_q(T)$ and hence $b(T)$ vanish identically. Here we shall consider the case of extreme non-cylindrical symmetry, namely the basal-plane ferromagnet ($\theta' = \theta = \frac{1}{2}\pi$). A straightforward, but tedious, calculation of the second derivatives of $U(T)$, when the strains are kept constant, gives

$$U_{\theta\theta} \equiv \left.\frac{\partial^2 U}{\partial\theta^2}\right|_{\varphi;\, m_k,\, b_k} = NJ(1 - \Delta M - \tfrac{1}{2}b)[A_0(T) - B_0(T)]$$

$$U_{\varphi\varphi} \equiv \left.\frac{\partial^2 U}{\partial\varphi^2}\right|_{\theta;\, m_k,\, b_k} = NJ(1 - \Delta M + \tfrac{1}{2}b)[A_0(T) + B_0(T)] \tag{23}$$

at $\theta = \frac{1}{2}\pi$, within the approximations by which we have determined $A_0(T) \pm B_0(T)$ (equation 14). When the magnetization is along a symmetry direction ($\varphi = p\pi/6$), we can make use of (20) to calculate the macroscopic value for the energy gap (21) and it is easily seen to agree with $E_{q=0}$ deduced in the random-phase approximation, (11c). If $\varphi \neq p\pi/6$, then $U_{\varphi\varphi}$ may differ from $F_{\varphi\varphi}$, implying some limitation in the validity of (21). In principle, however, it is possible to extract these extra contributions to $F_{\varphi\varphi}$ from the macroscopic measurements, allowing also a determination of the energy-gap parameters (23) in this case.

The combination of (20) and (23) guarantees a disappearance of the energy gap when the magnetization is along a symmetry direction, in cases where where $F_{\theta\theta}$ or $F_{\varphi\varphi}$ are zero implying that the expression deduced for the energy gap, (equation 14), is in accordance with the Goldstone theorem.

By the inclusion of all possible single-ion terms in (4) and by allowing ΔM and b to depend on (θ, φ), the above result is a generalization of that obtained by Brooks (1970). The occurrence of the $(1 \pm \frac{1}{2}b)$ factors in (23), which are not present in the corresponding expressions deduced by Brooks, is connected to a slightly different definition of the two correlation functions, ΔM and b. In fact, we shall eliminate these factors from (23) by defining a new pair of energy parameters:

$$A^0_q(T) \pm B^0_q(T) = (1 \pm \tfrac{1}{2}b)[A_q(T) \pm B_q(T)], \tag{24}$$

where $E_q(T)$ is given by (9b) also in terms of the new parameters. Besides the removal of the $(1 \pm \frac{1}{2}b)$ factors in (23) when $A^0_0(T) \pm B^0_0(T)$ are introduced instead of $A_0(T) \pm B_0(T)$, these new energy parameters have further advantages. The two-ion term, $-J[\mathscr{J}(0) - \mathscr{J}(\boldsymbol{q})]\ b$, in (8$c$) disappears in the corresponding expression for $B^0_q(T)$, and more important, the Zeeman Hamiltonian contributes only to $A^0_q(T)$. This gives the trans-

formation (24) a physical meaning, as $A_q^0(T) \pm B_q^0(T)$ are the parameters determined from the field dependence of the spin-wave energies (Jensen *et al* 1975). If we define ΔM^0 and b^0 as the parameters determined by (10) when $A_q(T)$ and $B_q(T)$ are replaced by $A_q^0(T)$ and $B_q^0(T)$, we have

$$\Delta M = \Delta M^0 + \tfrac{1}{2}(b^0)^2, \qquad b = b^0(1 + \tfrac{1}{4}J^{-1} + \tfrac{1}{2}\Delta M^0). \tag{25}$$

The static and dynamic energy-gap parameters deduced in Tb seem to agree satisfactorily (Houmann *et al* 1975) with the exception of the six-fold basal-plane anisotropy of the unstrained lattice. A non-zero value of b does not introduce any difference between the static and dynamic energy-gap parameters (at least not to first order in ΔM and b), meaning that b is not essential for explaining the appearance of such a difference. It has to be a dynamic effect due to either the coupling to the conduction electrons or a breakdown of the simple Hartree–Fock/random-phase approximations.

In the rare-earth metals, the coupling between the angular moments on different sites, (equation 1), is established via the spins of the conduction electrons which are polarized and hence contribute to the total magnetization. This may be included by defining an effective g-factor, $g_{\text{eff}} = g + \Delta g$, entering (3) and (22). Further, the interaction between the magnons and the conduction electrons causes a change of the magnon energies. In the random-phase approximation, the energy gap at zero wavevector is determined by

$$E'_{q=0} = E_{q=0}/(1 + \tfrac{1}{2}\Delta g) \tag{26}$$

(see eg Giovannini *et al* 1966; in (26) the relaxation of the conduction electrons is neglected). Notice that equation (21) leads to the same result only if $g = 2$ (S-state ion). The presence of anisotropic two-ion couplings may strongly enhance the modification appearing in (26). The dynamic corrections to the random-phase decoupled spin waves (the off-diagonal terms in (6)) are expected to behave in a similar fashion, being proportional to the energy of the normal mode considered. The difference between the energy-gap parameters deduced in Tb may be accounted for by the modification introduced by the conduction electrons alone, as is shown by an order-of-magnitude estimate (Houmann *et al* 1975).

The scaling of the dynamic corrections to the energy of the normal modes, arising from either of the two mechanisms, implies that they vanish for the magnetic contributions (Jensen 1971) to the long-wavelength phonons ($\approx$ zero energy). Within the present approximations, we find that the 'soft mode' behaviour expected for the transverse sound waves (which have their polarization vector in the basal plane and their propagation vector parallel to an applied field along the hard planar axis) is exhaustively described by the static magnetostriction parameters (C and A) and the value of the critical field (proper account has to be taken on the effects of the magnetic dipolar field).

4. Renormalization of the static anisotropy parameters

In this section we shall discuss the renormalization of anisotropy parameters as deduced from (15). If b is non-zero, then the Stevens operator expansion, (4*a*), and the usual definition of the magnetic anisotropy in terms of spherical harmonics for example is not directly related, as discussed in I and by Brooks and Egami (1973). Here we shall consider only the two combinations of anisotropy parameters, $U_{\theta\theta}$ and $U_{\varphi\varphi}$, which are relevant in determining the static value of the energy gap in the case of a basal-plane ferromagnet. For simplicity, we include only the two most important terms of the

magnetoelastic Hamiltonian, $\mathscr{H}_{\rm me}$ in (4*a*). These are the $l = 2$, B_2^2 contribution to C and the $l = 4$, B_4^4 contribution to A (the γ-strain parameters C and A are those defined in, for example, Houmann *et al* 1975). Proceeding as in I, we deduce the following expressions for $U_{\theta\theta}$ and $U_{\varphi\varphi}$ at $\theta = \pi/2$:

$$N^{-1}U_{\theta\theta} = N^{-1}F_{\theta\theta} = 6\tilde{B}_{20}^{-} - 60\tilde{B}_{40}^{-} + 210\tilde{B}_{60}^{-} - 6\tilde{B}_{66}^{-}\cos 6\varphi + 2c^{\gamma}\tilde{B}_{22}^{-}\tilde{B}_{22}^{+} + c^{\gamma}\tilde{B}_{44}^{-}\tilde{B}_{44}^{+} - c^{\gamma}(2\tilde{B}_{44}^{-}\tilde{B}_{22}^{+} + \tilde{B}_{22}^{-}\tilde{B}_{44}^{+})\cos 6\varphi$$

$$N^{-1}U_{\varphi\varphi} = 4c^{\gamma}(\tilde{B}_{22}^{+})^2 + 4c^{\gamma}(\tilde{B}_{44}^{+})^2 - 10c^{\gamma}\tilde{B}_{22}^{+}\tilde{B}_{44}^{+}\cos 6\varphi - 36\tilde{B}_{66}^{+}\cos 6\varphi, \tag{27b}$$

where we used the following notation for the magnetostriction parameters:

$$C(T) = \tilde{B}_{22}^{+}, \qquad A(T) = \tilde{B}_{44}^{+}, \qquad c^{\gamma} = 4c_{66}/N. \tag{27c}$$

Based on the calculation by Callen and Callen (1965, 1966), we neglect the small kinematic correction appearing as the J^{-1} factors in (15); and we further interpret (15) as the first terms appearing in an expansion of the following power laws:

$$\begin{aligned}\tilde{B}_{l0}^{-} &= B_l^0 S_l \sigma^{\alpha_l}(1 + \tilde{b})^{\alpha_l - 2}(1 + \tilde{b}^2)^{3 - \alpha_l/2},\\ \tilde{B}_{ll}^{-} &= B_l^l S_l \sigma^{\alpha_l}(1 + \tilde{b})^{\gamma(l,1)}(1 + \tilde{b}^2)^{\gamma(l,2)}\\ \tilde{B}_{ll}^{+} &= B_l^l S_l \sigma^{\alpha_l}(1 + \tilde{b})^{-\alpha_l - 1}(1 + \tilde{b}^2)^{\alpha_l - 1/2}.\end{aligned} \tag{28}$$

The exponents $\gamma(l, 1)$ and $\gamma(l, 2)$ are given in table 1, and $\tilde{b}$ is defined

$$\tilde{b} = b\sigma^{-\frac{3}{2}}. \tag{29}$$

Except for the higher-order corrections, the replacement of b by $\tilde{b}$ and the occurrence of the $(1 + \tilde{b}^2)$ terms, these are the results also deduced by Brooks and Egami (1973). The identification of power laws from the finite expansion (15) is not unique, however; (28) agrees with the low-temperature result obtained by Callen and Callen (1965, 1966) in the case where b can be neglected. At higher temperatures, Callen and Callen deduced a substantial kinematic correction to the $l(l + 1)/2$ power law, such that σ^{α_l} is replaced by σ^l in the limit of $\sigma \to 0$. The σ^{α_l} dependence in (28) can be expected to be valid only at temperatures satisfying

$$\Delta M(T) = 1 - \sigma(T) \ll 2/l, \tag{30a}$$

corresponding to a range for which l times the population of the boson levels are much smaller than the number of $(2J + 1)$ physical states. It is not possible to include all the b^2 terms of (15) in an expansion of a simple power law in $(1 \pm \tilde{b})$, as is shown in (28) by the appearance of terms in powers of $(1 + \tilde{b}^2)$. If we require this correction to be unimportant by

$$\tilde{b}(T) \ll 1/l, \tag{30b}$$

Table 1. The exponents $\gamma(l, 1)$ and $\gamma(l, 2)$ appearing in equation (28).

l	$\gamma(l, 1)$	$\gamma(l, 2)$
2	1	$\frac{3}{2}$
4	0	18
6	−5	$\frac{105}{2}$

then we may hope that this is the condition by which higher-order corrections in terms of powers of $(1 + \tilde{b}^n)$, $n \geqslant 3$, in (28) can also be neglected.

The characteristic correlation functions, $\Delta M^0(T)$ and $b^0(T)$, at zero temperature and as functions of temperature, have been calculated in Gd, Tb and Dy by Lindgård and Danielsen in I. Using their values in the case of Tb, we derived the zero-temperature values, $C_{lm}^{\pm}$, and effective power laws, $\beta_{lm}^{\pm}$, of the parameters in (28) defined by

$$\tilde{B}_{lm}^{\pm} = B_l^m S_l C_{lm}^{\pm} \tilde{\sigma}^{\beta_{lm}^{\pm}} \tag{31}$$

and given in table 2, where we have included the small corrections of $b(T)$ and $\Delta M(0)$ from (25). $\tilde{\sigma}(T)$ is the effective relative magnetization

$$\tilde{\sigma} = \tilde{\sigma}(T) = M(T)/M(0) \cong \sigma + \Delta M(0). \tag{32}$$

Table 2 shows that the deviations from the $l(l + 1)/2$ power law are not very dramatic, in spite of the large value of $b(T)$ used in the calculations. We remark that the terms

Table 2. The zero temperature values, $C_{lm}^{\pm}$, and effective power laws, $\beta_{lm}^{\pm}$, of the static anisotropy parameters $\tilde{B}_{lm}^{\pm}$. In the calculations we use $\Delta M(T) = 0{\cdot}0064 + [1 - \tilde{\sigma}(T)]$ and $b(T) = 0{\cdot}0323 + 0{\cdot}35[1 - \tilde{\sigma}(T)]$, corresponding to $\Delta M^0(T)$ and $b^0(T)$ deduced by Lindgård and Danielsen (1975) in the case of Tb. The effective power laws, $\beta_{lm}^{\pm}$, only vary by a few percent in the range $1 > \tilde{\sigma} > 0{\cdot}8$, and they are comparable with the classical $\alpha_l = l(l + 1)/2$ power law.

l	m	$\pm$	$C_{lm}^{\pm}$	$\beta_{lm}^{\pm}$	α_l
2	0, 2	−	1·01	2·6	3
4	0	−	1·21	7·0	10
4	4	−	0·96	9·6	10
6	0	−	1·59	13·9	21
6	6	−	0·79	21·6	21
2	2	+	0·95	3·3	3
4	4	+	0·78	12·3	10
6	6	+	0·54	26·8	21

proportional to b^2 in the expansions of the power laws in (28) cannot be neglected, not even in the limit of zero temperature, because $b(0) \neq 0$ and the coefficients to b^2 are of the order of α_l^2.

The large hexagonal anisotropy present in Dy may cause a significant φ-dependence of ΔM and b. When an external field is applied along a hard planar axis in Dy or Tb, then the angle δ between the directions of the magnetization and the applied field is determined by the equilibrium condition (19):

$$Ng\mu_B J\sigma H \sin\delta - \tfrac{1}{6}K_6^6(\delta)\sin 6\delta = 0, \tag{33a}$$

where K_6^6 depends on the angle δ if ΔM or b are φ-dependent:

$$N^{-1}K_6^6(\varphi) = \left|36\tilde{B}_{66}^{+}(\varphi) + 18c^{\gamma}\tilde{B}_{22}^{+}(\varphi)\tilde{B}_{44}^{+}(\varphi)\right|. \tag{33b}$$

The parameter K_6^6 deduced from magnetization measurements (Feron *et al* 1970) is mostly determined by the value of the critical field, H_c, at which δ becomes zero. This transition is of second order only as long as

$$K_6^6(\text{hard}) > \sim \tfrac{3}{5}K_6^6(\text{easy}), \tag{33c}$$

Table 3. A_0^0 and B_0^0 are the energy-gap parameters of Dy at zero temperature (in units of meV). The two parameters are derived using the known value of $E_{q=0}$ (easy) and the anisotropy measurements by Feron *et al* (1970). The calculation performed by Lindgård and Danielsen (1975) of $\Delta M^0(0)$ and $b^0(0)$ in Dy (using a different value of B_0^0) is then utilized in a variational calculation of $\Delta M(0)$ and $b(0)$ given in the table. Using these parameters, we determined the relative zero-temperature values, C_{66}^+, of K_6^6 in the two cases in which the magnetization is along an easy and a hard direction.

	A_0	B_0^0	$\Delta M(0)$	$b(0)$	C_{66}^+
Easy	3·68	−1·80	0·0021	0·0168	0·746
Hard	3·38	−2·96	0·0068	0·0305	0·554

assuming the φ-dependent part of K_6^6 to be proportional to cos 6φ only. If K_6^6 is considered to be independent of φ, then the energy gap in Dy at zero temperature and zero field is calculated to be 2·6 ± 0·4 meV (Brooks and Egami 1974, Houmann *et al* 1975) using the static anisotropy parameters determined by Feron *et al* (1970). Introducing the change of K_6^6 estimated in table 3, $K_6^6(\text{easy}) \cong 1{\cdot}35K_6^6(\text{hard})$ where $K_6^6(\text{hard})$ is the parameter determined by the magnetization measurements, we find $E_{q=0}(\text{easy}) \cong 2{\cdot}9$ meV. This number is in fair agreement with the experimental energy gap of 3·0–3·2 meV (Marsh and Sievers 1969, Nicklow and Wakabayashi 1972), supporting the proposal of Egami (1972) of the importance of zero-point effects in Dy. When the temperature is increased, the relative difference between K_6^6(easy) and K_6^6(hard) should decrease, implying that the energy gap deduced from the static measurements, without correcting for the φ-dependence of K_6^6, should approach the experimental value. This is the case as shown in figure 1. ΔM and b, and hence K_6^6, depend on the strength of the applied field. Corresponding to the estimate given in table 3, we find that K_6^6(easy) should increase by 3·1 % per 100 kOe

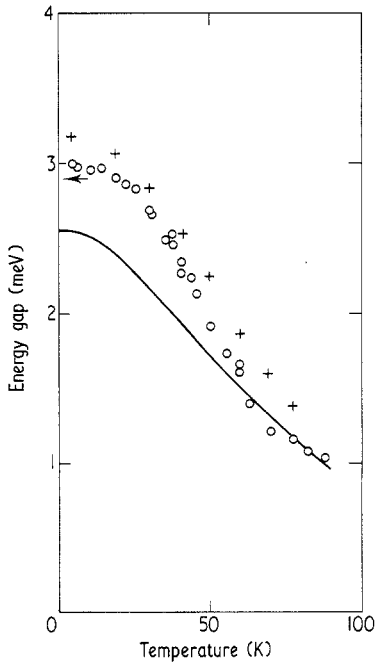

Figure 1. The spin-wave energy gap at $q = 0$ as a function of temperature in Dy, measured by infrared resonance (○, Marsh and Sievers 1969) and by inelastic neutron scattering (+, Nicklow and Wakabayashi 1972). The full curve displays the result obtained from the magnetization measurements (Feron *et al* 1970) when neglecting a φ-dependence of K_6^6. The arrow points at the value deduced at zero temperature when the estimated zero point change of K_6^6, given in table 3, is included.

and K_6^6(hard) by 7·5 % per 100 kOe, both at zero temperature, which numbers are somewhat smaller than those estimated by Brooks and Egami (1974).

At low temperatures, $\tilde{\sigma}(T) > 0{\cdot}9$, the parameters $\Delta M(T)$ and $b(T)$ used in the tables 2 and 3 fulfill the conditions (30) for all values of $l \leqslant 6$. A much more severe limitation of the numerical results above is raised by the assumption of $B_{\boldsymbol{q}}^0(T)$ being independent of $\boldsymbol{q}$ in the calculation of $b(T)$. The measurements of $B_{\boldsymbol{q}}^0(T)$ in Tb as a function of $\boldsymbol{q}$ parallel with the c direction (Jensen *et al* 1975) have revealed a strong $\boldsymbol{q}$-dependence of $B_{\boldsymbol{q}}^0(T)$ which is interpreted in terms of large anisotropic two-ion couplings. The $\boldsymbol{q}$-dependent contribution to $B_{\boldsymbol{q}}^0(T)$ arising from the isotropic two-ion coupling (the last term in (8c)) is entirely negligible in comparison with $B_0^0(T)$ (being smaller by a factor of 10 at least). The observed $\boldsymbol{q}$-dependence of $B_{\boldsymbol{q}}^0(T)$ implies that the values of $\Delta M(0)$ and $b(T)$ used in the tables 2 and 3 are subjected to large uncertainties. More reliable values may only be derived when $B_{\boldsymbol{q}}^0(T)$ has been determined in all symmetry directions.

5. Conclusion

In the spin-wave theory of an anisotropic ferromagnet ($B_{\boldsymbol{q}} \neq 0$), the application of the Holstein–Primakoff transformation implies that single-ion contributions to the spin-wave energies are transferred to the two-ion part of the Hamiltonian. We included these contributions to the energy of the uniform spin-wave mode in a random-phase calculation and obtained essentially the result deduced by Brooks (1970) using an alternative spin operator technique. The advantages achieved by the use of the method of Lindgård and Danielsen (1974, 1975) of a generalized Holstein–Primakoff transformation, in comparison with the one used by Brooks (1970) and Brooks and Egami (1973), are a more precise definition of the characteristic correlation functions, $\Delta M(T)$ and $b(T)$, in terms of the spin-wave Hamiltonian, and the more systematic way in which kinematics effects are encountered.

We presented numerical results for the renormalization of anisotropy parameters in Tb and Dy which were based on the calculations of $\Delta M(T)$ and $b(T)$ by Lindgård and Danielsen (1975). Our results showed the effects of $b(T)$ to be less drastic than reported in the previous papers. We pointed out that the numerical results can only be of a qualitative nature until the spin-wave parameter, $B_{\boldsymbol{q}}^0(T)$, has been determined as a function of the wavevector along all the symmetry directions.

Acknowledgments

We should like to thank A P Young for valuable suggestions and a critical reading of the manuscript. Helpful discussions with D Melville and R J Elliott are gratefully acknowledged.

References

Brooks M S S, Goodings D A and Ralph H I 1968 *J. Phys.* C: *Solid St. Phys.* **1** 132–45
Brooks M S S 1970 *Phys. Rev.* B **1** 2257–64
Brooks M S S and Egami T 1973 *J. Phys.* C: *Solid St. Phys.* **6** 513–31, 3719–36
Brooks M S S and Egami T 1974 *J. Phys.* C: *Solid St. Phys.* **7** 979–94
Callen E and Callen H B 1965 *Phys. Rev.* **139** A455–71

Callen H B and Callen E 1966 *J. Phys. Chem. Solids* **27** 1271–85
Egami T 1972 *J. Phys.* C: *Solid St. Phys.* **5** L85–8
Feron J L, Huy G and Pauthenet R 1970 *Les Elements des Terres Rares II* (CRNS No 180 Paris) pp 19–30
Giovannini B, Peter M and Koide S 1966 *Phys. Rev.* **149** 251–6
Goodings D A and Southern B 1971 *Can. J. Phys.* **49** 1139–61
Houmann J G, Jensen J and Touborg P 1975 *Phys. Rev.* to be published
Jensen J 1971 *Risø Report No 252* Danish Atomic Energy Commission
Jensen J, Houmann J G and Møller H B 1975 *Phys. Rev.* to be published
Lindgård P A and Danielsen O 1974 *J. Phys.* C: *Solid St. Phys.* **7** 1523–35
—— 1975 *Phys. Rev.* B **11** 351–62
Marsh H S and Sievers A J 1969 *J. Appl. Phys.* **40** 1563–4
Nicklow R M and Wakabayashi N 1972 *Neutron Inelastic Scattering* (Vienna: IAEA) pp 611–7
Rhyne J J and Clark A E 1967 *J. Appl. Phys.* **38** 1379–80
Smith J and Beljers H G 1955 *Phillips Res. Rep.* **10** 113–30