PHYSICAL REVIEW B VOLUME 11, NUMBER 1 1 JANUARY 1975

Theory of magnetic properties of heavy rare-earth metals: Temperature dependence of magnetization, anisotropy, and resonance energy

Per-Anker Lindgård and Oluf Danielsen

Danish Atomic Energy Commission, Research Establishment Risø, DK-4000 Roskilde, Denmark

(Received 4 February 1974)

The contributions to the macroscopic-anisotropy constants and resonance energy from crystal-field anisotropy, magnetoelastic effects in the frozen and flexible lattice model, and two-ion interactions have been found for all terms allowed in a crystal of hexagonal symmetry. The temperature dependence is expressed as expansions of thermal averages of the Stevens operators $\langle O_l^m \rangle$. A systematic spin-wave theory, renormalized in the Hartree-Fock approximation, is developed and used to find the temperature dependence of the Stevens operators and the resonance energy in terms of the magnetization-deviation parameter $\Delta M(T)$ and the parameter $b(T)$, which characterizes the nonsphericity of the moment precession. Significant deviations from the classical $l(l+1)/2$ temperature law are found. The inclusion of $b(T)$ gives rise to important reinterpretations of the contributions to the resonance energy. Numerical results are given for the magnetization agreeing with experiment for Gd, Tb, and Dy. For Tb and Dy the zero-point deviations were found to be $0.05\mu_B$ and $0.08\mu_B$, respectively, and the ratio $[b(T) - b(0)]/[\Delta M(T) - \Delta M(0)]$ is approximately 1/3 for all temperatures below 100 K. This gives rise to large corrections of the results of previous theories. Tables of these corrections are given for the resonance energy and the macroscopic-anisotropy constants. For planar anisotropy the temperature renormalization is reduced for the axial anisotropy and increased for the in-plane anisotropy.

I. INTRODUCTION

The magnetic properties of the heavy rare-earth metals have been extensively studied experimentally in the last decade. A comprehensive review is available.[1] Experiments have shown that the magnetic properties are a result of an intricate interplay of many forces of comparable magnitude. The dominant interaction is the indirect exchange interaction, and of importance are also crystal-field anisotropy, magnetoelastic effects, and possibly also two-ion anisotropy. In general, however, the experiments have been analyzed on the basis of simple classical and molecular-field-type theories which can only be expected to account for the major features correctly. The molecular-field-type theory of Callen and Callen[2] for the temperature dependence of the macroscopic anisotropy and magnetostriction has been a very successful first-order theory. The classical (i.e., the infinite-angular-momentum J limit) spin-wave theory of Cooper,[3] which includes a majority of the physically important effects and with a renormalization in analogy with classical expressions for the resonance energy, has also been very useful in structuring the problems. The quantum-mechanical corrections for finite J are of importance and have been discussed by Lindgård *et al.*[4] and later by Brooks and Goodings and co-workers.[5] They have also studied the effect of the nonspherical precession of the angular momentum under the influence of anisotropy.[5,16] However the spin-wave treatment of the anisotropy terms is complicated and was not done systematically. A basis for a systematic treatment has been developed by Lindgård and Danielsen[6] by a generalization of the Holstein-Primakoff transformation which can be applied to tensor operators of arbitrary rank. Using this transformation it is simple to treat all the various terms relevant for the heavy rare-earth metals. For simplicity and as a first correction to the molecular-field-type theories, the dynamical interaction is treated in the Hartree-Fock approximation. Although this approximation does not exactly reproduce the results by Dyson[21] for a Heisenberg ferromagnet it has proved to compare well with experiments for various test systems as for instance the planar antiferromagnet $NiCl_2$.[22] It is the aim of this paper to carry out this calculation for the heavy rare-earth metals and show that significant corrections to the first-order theories are obtained. The results are also valid for other magnetic systems of hexagonal symmetry.

It is well known that the spin-wave theory using the Holstein Primakoff transformation is only valid at low temperatures, where the number of thermally excited spin waves is small.[14] At higher temperatures the neglected kinematical interaction, which prevents more than $2J$ excitations of a single ion, becomes important. However, for the heavy rare-earth metals the J values are large ($J \geq 3.5$) and the presence of large energy gaps in the spin-wave spectra reduces the number of excitations. A renormalized spin-wave theory, which only treats two-magnon interactions, is therefore expected to work well for this group of materials.

In Sec. II the most important features of the heavy rare-earth metals are summarized. Section

III gives the relation between the macroscopic and the microscopic anisotropy parameters. In Sec. IV the spin-wave theory, renormalized in the Hartree-Fock approximation, is developed and the results are given for the temperature dependence of the anisotropy parameters and also for the resonance energy (the spin-wave energy gap). Finally, in Sec. V the magnitude of these effects are investigated by applying the theory to gadolinium, terbium, and dysprosium. Section VI is a short summary. The Appendix gives an explicit discussion of the planar anisotropy case, including magnetoelastic effects.

II. BASIC INTERATIONS IN HEAVY RARE-EARTH METALS

The crystal structure of the heavy rare-earth metals is close to the hexagonal-close-packed structure. In the magnetically ordered phase it is distorted by magnetostriction.[1] We shall consider the effect of the isotropic exchange interaction, the single-ion anisotropy, the single-ion magnetostriction, and an external applied field. Two-ion anisotropy is included in a general phenomenological form. The Hamiltonian therefore contains the following terms:

$$H = H_{\mathrm{ex}} + H_{1\ \mathrm{an}} + H_{\mathrm{me}} + H_{\mathrm{el}} + H_{\mathrm{Zee}} + H_{2\ \mathrm{an}} \ . \tag{1}$$

As the hexagonal-close-packed structure can be divided into two interpenetrating Bravais sublattices, the isotropic exchange term takes the form

$$\begin{aligned} H_{\mathrm{ex}} = &-\sum_{f>g} 2\mathcal{J}(\vec{\mathrm{R}}_{fg})\, \vec{\mathrm{J}}_f \cdot \vec{\mathrm{J}}_g \\ &-\sum_{f>f'} 2\mathcal{J}'(\vec{\mathrm{R}}_{ff'})\, \vec{\mathrm{J}}_f \cdot \vec{\mathrm{J}}_{f'} \ . \end{aligned} \tag{2}$$

$\mathcal{J}(\vec{\mathrm{R}}_{fg})$ is the exchange interaction between sites f and g within the same sublattice, and $\mathcal{J}'(\vec{\mathrm{R}}_{ff'})$ is the interaction between f and f' in different sublattices.

Cooper, Elliott, Nettel, and Suhl[7] have given the single-ion anisotropy term per ion, with the quantization axis along the hexagonal c axis as follows:

$$H_{1\ \mathrm{an}} = B_2^0 O_2^0(c) + B_4^0 O_4^0(c) + B_6^0 O_6^0(c) + B_6^6 O_6^6(c) \ . \tag{3}$$

The B_l^m coefficients are the crystal-field parameters and $O_l^m(c)$ are the cosine-type Stevens-operator equivalents, defined by Elliott and Stevens.[8]

Expanded in terms of the irreducible strains of the hexagonal-close-packed lattice, the single-ion magnetostriction per ion is (Callen and Callen[2])

$$\begin{aligned} H_{\mathrm{me}} = &-\sum_{\Gamma, i} B^{\Gamma,i} S^{\Gamma,i} \epsilon^{\Gamma,i} \\ = &-\sum_{\substack{L \\ M=0,6}} (B_{LM}^{\alpha,1}\epsilon^{\alpha,1} + B_{LM}^{\alpha,2}\epsilon^{\alpha,2})\, O_L^M(c) \\ &+\sum_L \{B_{L2}^{\gamma}[\epsilon_1^{\gamma}\, O_L^2(c) + \epsilon_2^{\gamma}\, O_L^2(s)] \\ &+ B_{L4}^{\gamma}[\epsilon_1^{\gamma}\, O_L^4(c) - \epsilon_2^{\gamma}\, O_L^4(s)] \\ &+ B_{L1}^{\epsilon}[\epsilon_1^{\epsilon}\, O_L^1(c) + \epsilon_2^{\epsilon}\, O_L^1(s)]\} \\ &+ B_{65}^{\epsilon}[\epsilon_1^{\epsilon}\, O_6^5(c) - \epsilon_2^{\epsilon}\, O_6^5(s)] \ , \end{aligned} \tag{4}$$

where the B's are the magnetoelastic constants, and $L = 2$, 4, and 6; $S^{\Gamma,i}$ are the appropriate sums of Stevens operators; $\epsilon^{\Gamma,i}$ are the irreducible strains. $O_l^m(s)$ are the sine-type Stevens operators. The elastic term per ion is

$$\begin{aligned} H_{\mathrm{el}} &= \frac{1}{2}\sum_{\substack{\Gamma \\ i,j}} c_{ij}^{\Gamma}\epsilon^{\Gamma,i}\epsilon^{\Gamma,j} \\ &= \tfrac{1}{2}c_{11}^{\alpha}(\epsilon^{\alpha,1})^2 + c_{12}^{\alpha}\epsilon^{\alpha,1}\epsilon^{\alpha,2} + \tfrac{1}{2}c_{22}^{\alpha}(\epsilon^{\alpha,2})^2 \\ &\quad + \tfrac{1}{2}c^{\gamma}[(\epsilon_1^{\gamma})^2 + (\epsilon_2^{\gamma})^2] + \tfrac{1}{2}c^{\epsilon}[(\epsilon_1^{\epsilon})^2 + (\epsilon_2^{\epsilon})^2] \ ; \end{aligned} \tag{5}$$

c_{ij}^{Γ} are the elastic constants. The irreducible strains in terms of the Cartesian strains are given by Callen and Callen[2]:

$$\begin{aligned} &\epsilon^{\alpha,1} = \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz} \\ &\epsilon^{\alpha,2} = (1/2\sqrt{3})(3\epsilon_{zz} - \epsilon^{\alpha,1}) \ , \\ &\epsilon_1^{\gamma} = \tfrac{1}{2}(\epsilon_{xx} - \epsilon_{yy}) \ , \\ &\epsilon_2^{\gamma} = \epsilon_{xy}, \qquad \epsilon_1^{\epsilon} = \epsilon_{yz}, \qquad \epsilon_2^{\epsilon} = \epsilon_{xz} \ . \end{aligned} \tag{6}$$

When the lattice is in equilibrium with respect to the strain, we find, by differentiation of $H_{\mathrm{el}} + H_{\mathrm{me}}$ with respect to $\epsilon^{\Gamma,i}$, the equilibrium strains

$$\begin{aligned} &\epsilon^{\alpha,i} = \frac{1}{c_{ii}^{\alpha}c_{jj}^{\alpha} - (c_{ij}^{\alpha})^2} \sum_{\substack{L \\ M=0,6}} (c_{jj}^{\alpha}B_{LM}^{\alpha,i} - c_{ij}^{\alpha}B_{LM}^{\alpha,j})\, O_L^M(c) \ , \\ &\qquad\qquad (i,j) = (1,2) \text{ or } (2,1) \\ &\epsilon_n^{\gamma} = \frac{1}{c^{\gamma}}\sum_L \left[B_{L2}^{\gamma} O_L^2\binom{c}{s} \pm B_{L4}^{\gamma} O_L^4\binom{c}{s}\right], \qquad n = \begin{cases}1\\2\end{cases}, \\ &\epsilon_n^{\epsilon} = \frac{1}{c^{\epsilon}}\sum_L \left[B_{L1}^{\epsilon} O_L^1\binom{c}{s} \pm B_{65}^{\epsilon} O_6^5\binom{c}{s}\right], \qquad n = \begin{cases}1\\2\end{cases}. \end{aligned} \tag{7}$$

The strains in thermodynamic equilibrium $\langle\epsilon^{\Gamma,i}\rangle$ are found by taking the thermodynamic equilibrium value of the Stevens operators. By substitution in (4) and (5) we find in equilibrium with respect to strain

$$H_{\mathrm{el}} + H_{\mathrm{me}}\big|_{\mathrm{eq}} = -H_{\mathrm{el}} \ . \tag{8}$$

An externally applied magnetic field $\vec{\mathrm{H}}$ contributes with a term in the Hamiltonian

$$H_{\mathrm{Zee}} = -g\mu_B \sum_f \vec{\mathrm{H}} \cdot \vec{\mathrm{J}}_f \ ,$$

where g is the Landé factor[1] and μ_B is the Bohr magneton.

The two-ion anisotropy term can generally be represented by

$$H_{2\ \mathrm{an}} = -\sum_{\substack{l'm'i' \\ l''m''i'' \\ fg}} K_{l'm'i'}^{l''m''i''}(f,g)\, O_{l'}^{m'}(i')_f\, O_{l''}^{m''}(i'')_g \ , \tag{9}$$

where the constants of interaction between sites f and g are $K_{l''m''i''}^{l'm'i'}(f,g)$.

III. RELATION BETWEEN MACROSCOPIC AND MICROSCOPIC ANISOTROPY PARAMETERS

The macroscopic anisotropy parameters are the coefficients in a symmetry-determined expansion of the free energy in terms dependent on the direction of the magnetization specified by the polar coordinates (θ, ϕ) relative to the crystal axis. The microscopic anisotropy parameters are terms of various physical origins which enter in the Hamiltonian for the system. The two sets of parameters are related by the expression

$$F(\theta, \phi) = -k_B T \ln \mathrm{Tr}(e^{-\beta H(\theta, \phi)}) , \tag{10}$$

where k_B is Boltzmann's constant, T is the temperature, and $\beta = 1/k_B T$. $H(\theta, \phi)$ is the Hamiltonian in a representation in which the quantization axis is along the magnetization direction. For hexagonal symmetry the free energy is generally assumed to be[1]

$$F(\theta, \phi)_{\text{hex}} = K_0(T) + K_1(T)\sin^2\theta + K_2(T)\sin^4\theta + K_3(T)\sin^6\theta + K_4(T)\sin^6\theta\cos 6\phi , \tag{11}$$

where θ is the angle of the magnetization relative to the hexagonal axis and ϕ the angle in the basal plane relative to the direction to the nearest-neighbor atom.

If the hexagonal symmetry is distorted by the presence of strains, other terms of the appropriate symmetry will occur. In the following we shall, however, only consider the hexagonal terms.

A simple relation between the free energy and the Hamiltonian is obtained by differentiation with respect to θ and ϕ:

$$\begin{aligned} \frac{\partial}{\partial\theta} F(\theta, \phi)\Big|_{\theta_0, \phi_0} &= \left\langle \frac{\partial}{\partial\theta} H(\theta, \phi) \right\rangle\Big|_{\theta_0, \phi_0} , \\ \frac{\partial}{\partial\phi} F(\theta, \phi)\Big|_{\theta_0, \phi_0} &= \left\langle \frac{\partial}{\partial\phi} H(\theta, \phi) \right\rangle\Big|_{\theta_0, \phi_0} , \end{aligned} \tag{12}$$

$$\begin{aligned} \frac{\partial}{\partial\phi} F(\theta, \phi)_{\text{hex}} &= K_1(T)\sin 2\theta + 2K_2(T)\sin^2\theta\sin 2\theta \\ &\quad + 3K_3(T)\sin^4\theta\sin 2\theta + 3K_4(T)\sin^4\theta \\ &\quad \times \sin 2\theta\cos 6\phi , \\ \frac{\partial}{\partial\phi} F(\theta, \phi)_{\text{hex}} &= -6K_4(T)\sin^6\theta\sin 6\phi . \end{aligned} \tag{13}$$

The angular dependence of the Hamiltonian $H(\theta, \phi)$ is found by rotating each term in the Hamiltonian to a representation with the quantization axis along a direction specified by θ and ϕ. Such rotations of Stevens operators have been treated in detail by Danielsen and Lindgård.[9] The rotated operators are linear combinations of Stevens operators with coefficients dependent on θ and ϕ. The coefficients are differentiated with respect to θ and ϕ, and, since (12) must hold for any set of (θ_0, ϕ_0), the right- and left-hand sides are equated term by term. Following this procedure we find for the contribution from the single-ion anisotropy terms

$$\begin{aligned} K_1(T) &= -\tfrac{3}{2}B_2^0(\langle O_2^0\rangle + \langle O_2^2\rangle) - 5B_4^0(\langle O_4^0\rangle + 3\langle O_4^2\rangle) \\ &\quad - \tfrac{21}{2}B_6^0(\langle O_6^0\rangle + 5\langle O_6^2\rangle) , \\ K_2(T) &= \tfrac{35}{8}B_4^0(\langle O_4^0\rangle + 4\langle O_4^2\rangle + \langle O_4^4\rangle) \\ &\quad + \tfrac{63}{8}B_6^0(\langle O_6^0\rangle + 20\langle O_6^2\rangle + 5\langle O_6^4\rangle) , \\ K_3(T) &+ -\tfrac{231}{16}B_6^0(\langle O_6^0\rangle + \tfrac{15}{2}\langle O_6^2\rangle + 3\langle O_6^4\rangle + \tfrac{1}{2}\langle O_6^6\rangle) , \\ K_4(T) &= \tfrac{1}{16}B_6^6(\langle O_6^0\rangle + \tfrac{15}{2}\langle O_6^2\rangle + 3\langle O_6^4\rangle + \tfrac{1}{2}\langle O_6^6\rangle) \\ &= -\tfrac{1}{24}K_3(T), \quad \text{for a hexagonal crystal field.} \end{aligned} \tag{14}$$

$\langle O_l^m\rangle$ are the average values of the cosine-type Stevens operators in a coordinate system with the z axis along the magnetization direction (θ_0, ϕ_0). It is somewhat difficult in general to assess the approximation involved in the method of determining the anisotropy constants $K_n(T)$ by means of (12) and (13). In the following section the problem will be discussed within the framework of the spin-wave theory.

The free energy for hexagonal symmetry is also conveniently written in an expansion of spherical harmonics as follows[1]:

$$F(\theta, \phi) = \kappa_0(T) + \kappa_2(T)Y_2^0(\theta, \phi) + \kappa_4(T)Y_4^0(\theta, \phi) + \kappa_6(T)Y_6^0(\theta, \phi) + \kappa_6^6(T)\sin^6\theta\cos 6\phi . \tag{15}$$

The so-called anisotropy coefficients $\kappa_l(T)$ are simply related to the anisotropy constants $K_n(T)$.[1] We find

$$\begin{aligned} \kappa_2(T) &= B_2^0(\langle O_2^0\rangle + \langle O_2^2\rangle) - \tfrac{10}{3}B_4^0(\langle O_4^2\rangle + (O_4^4\rangle) \\ &\quad - \tfrac{1}{2}B_6^0(5\langle O_6^2\rangle - 6\langle O_6^4\rangle - 11\langle O_6^6\rangle) , \\ \kappa_4(T) &= B_4^0(\langle O_4^0\rangle + 4\langle O_4^2\rangle + \langle O_4^4\rangle) \\ &\quad - \tfrac{9}{10}B_6^0(5\langle O_6^2\rangle + 8\langle O_6^4\rangle + 3\langle O_6^6\rangle) , \\ \kappa_6(T) &= B_6^0(\langle O_6^0\rangle + \tfrac{15}{2}\langle O_6^2\rangle + 3\langle O_6^4\rangle + \tfrac{1}{2}\langle O_6^6\rangle) , \\ \kappa_6^6(T) &= K_4(T) . \end{aligned} \tag{16}$$

The anisotropy coefficients $\kappa_l(T)$ are most directly related to the microscopic parameters B_l^0, since $\langle O_l^0\rangle \gg \langle O_l^{m\neq 0}\rangle$.

The treatment of the magnetoelastic terms in the Hamiltonian requires an additional consideration. Let us consider two physical extremes. In the frozen-lattice model the strains are fixed at the equilibrium values and independent of the actual direction of the moment (θ, ϕ). This model is likely to hold in the case of rapid moment precession, which occurs in the propagation of spin waves in anisotropic systems with a large spin-wave energy gap. In macroscopic measurements where the magnetization is slowly tilted to the polar coordi-

TABLE I. Coefficients in the reduction of a product of two Stevens operators

$$O_{l'}^{m'}(c)\,O_{l''}^{m''}(c)=\sum_{L,M}[l'm',\ l''m'',\ LM]\,O_L^M(c).$$

For a general angular momentum J the coefficient can be written as the sum $\sum_n C_n X^n$, where $X=J(J+1)$. Only the coefficients relevant for this paper are given, a complete table is given elsewhere (Ref. 19).

$l'm'$	$l''m''$	LM	C_0	C_1	C_2	C_3	C_4	C_5
20	20	20	$-2.1429\times10^{+0}$	5.7143×10^{-1}				
20	20	40	2.5714×10^{-1}					
21	21	20	-8.9286×10^{-2}	2.3810×10^{-2}				
21	21	40	-1.4286×10^{-2}					
22	22	20	7.1429×10^{-1}	-1.9048×10^{-1}				
22	22	40	1.4286×10^{-2}					
40	20	20	$1.7143\times10^{+1}$	$-1.3143\times10^{+1}$	$2.2857\times10^{+0}$			
40	20	40	$-7.4026\times10^{+0}$	5.1948×10^{-1}				
40	20	60	4.5455×10^{-1}					
41	21	20	7.1429×10^{-1}	-5.4762×10^{-1}	9.5238×10^{-2}			
41	21	40	-9.2532×10^{-2}	6.4935×10^{-3}				
41	21	60	-1.5152×10^{-2}					
42	22	20	$1.4286\times10^{+0}$	$-1.0952\times10^{+0}$	1.9048×10^{-1}			
42	22	40	$1.1104\times10^{+0}$	-7.7922×10^{-2}				
42	22	60	1.5152×10^{-2}					
44	22	66	5.0000×10^{-1}					
40	40	20	$-4.9351\times10^{+2}$	$4.1299\times10^{+2}$	$-9.2352\times10^{+1}$	$4.6176\times10^{+0}$		
40	40	40	$2.8537\times10^{+2}$	$-5.2759\times10^{+1}$	$1.2947\times10^{+0}$			
40	40	60	$-3.6970\times10^{+1}$	8.0808×10^{-1}				
41	41	20	$-1.0487\times10^{+1}$	$8.7760\times10^{+0}$	$-1.9625\times10^{+0}$	9.8124×10^{-2}		
41	41	40	$3.5672\times10^{+0}$	-6.5949×10^{-1}	1.6184×10^{-2}			
41	41	60	4.6212×10^{-2}	-1.0101×10^{-3}				
42	42	20	$-9.8701\times10^{+0}$	$8.2597\times10^{+0}$	$-1.8470\times10^{+0}$	9.2352×10^{-2}		
42	42	40	$-8.7198\times10^{+0}$	$1.6121\times10^{+0}$	-3.9560×10^{-2}			
42	42	60	$2.0333\times10^{+0}$	-4.4444×10^{-2}				
44	42	66	$1.2200\times10^{+1}$	-2.6667×10^{-1}				
44	44	20	$1.9740\times10^{+1}$	$-1.6519\times10^{+1}$	$3.6941\times10^{+0}$	-1.8470×10^{-1}		
44	44	40	$6.3417\times10^{+0}$	$-1.1724\times10^{+0}$	2.8771×10^{-2}			
44	44	60	2.1126×10^{-1}	-4.6176×10^{-3}				
60	20	40	$6.6084\times10^{+1}$	$-1.8566\times10^{+1}$	$1.2587\times10^{+0}$			
60	20	60	$-1.5655\times10^{+1}$	5.0909×10^{-1}				
61	21	40	$1.8357\times10^{+0}$	-5.1573×10^{-1}	3.4965×10^{-2}			
61	21	60	-9.3182×10^{-2}	3.0303×10^{-3}				
62	22	40	$2.9371\times10^{+0}$	-8.2517×10^{-1}	5.5944×10^{-2}			
62	22	60	$1.4909\times10^{+0}$	-4.8485×10^{-2}				
64	22	66	$4.1000\times10^{+0}$	-1.3333×10^{-1}				
65	21	66	$-1.0250\times10^{+0}$	3.3333×10^{-2}				
60	40	20	$4.4057\times10^{+3}$	$-4.6154\times10^{+3}$	$1.6203\times10^{+3}$	$-2.2937\times10^{+2}$	$1.1189\times10^{+1}$	
60	40	40	$-5.3748\times10^{+3}$	$1.6276\times10^{+3}$	$-1.3538\times10^{+2}$	$2.2378\times10^{+0}$		
60	40	60	$1.3566\times10^{+3}$	$-1.1470\times10^{+2}$	$1.1979\times10^{+0}$			
61	41	20	$7.3429\times10^{+1}$	$-7.6923\times10^{+1}$	$2.7005\times10^{+1}$	$-3.8228\times10^{+0}$	1.8648×10^{-1}	
61	41	40	$-6.7185\times10^{+1}$	$2.0344\times10^{+1}$	$-1.6923\times10^{+0}$	2.7972×10^{-2}		
61	41	60	$8.0749\times10^{+0}$	-6.8271×10^{-1}	7.1301×10^{-3}			
62	42	20	$1.1749\times10^{+2}$	$-1.2308\times10^{+2}$	$4.3207\times10^{+1}$	$-6.1166\times10^{+0}$	2.9837×10^{-1}	
62	42	60	$-3.5529\times10^{+1}$	$3.0039\times10^{+0}$	-3.1373×10^{-2}			
62	44	66	$1.0659\times10^{+2}$	$-9.0118\times10^{+0}$	9.4118×10^{-2}			
64	42	66	$3.9971\times10^{+2}$	$-3.3794\times10^{+1}$	3.5294×10^{-1}			
64	44	20	$4.1959\times10^{+1}$	$-4.3956\times10^{+1}$	$1.5431\times10^{+1}$	$-2.1845\times10^{+0}$	1.0656×10^{-1}	
64	44	40	$1.5357\times10^{+2}$	$-4.6501\times10^{+1}$	$3.8681\times10^{+0}$	-6.3936×10^{-2}		
64	44	60	$3.2299\times10^{+1}$	$-2.7308\times10^{+0}$	2.8520×10^{-2}			
65	41	66	$-6.6618\times10^{+1}$	$5.6324\times10^{+0}$	-5.8824×10^{-2}			
60	60	20	$-1.5172\times10^{+5}$	$1.6389\times10^{+5}$	$-6.0972\times10^{+4}$	$9.7142\times10^{+3}$	$-6.4224\times10^{+2}$	$1.2531\times10^{+1}$
60	60	40	$1.9723\times10^{+5}$	$-7.2086\times10^{+4}$	$8.6157\times10^{+3}$	$-3.6655\times10^{+2}$	$3.3172\times10^{+0}$	
60	60	60	$-6.6507\times10^{+4}$	$9.0599\times10^{+3}$	$-3.1703\times10^{+2}$	$1.8013\times10^{+0}$		
61	61	20	$-1.6772\times10^{+3}$	$1.8117\times10^{+3}$	$-6.7401\times10^{+2}$	$1.0739\times10^{+2}$	$-7.0996\times10^{+0}$	1.3853×10^{-1}
61	61	40	$1.7889\times10^{+3}$	$-6.5384\times10^{+2}$	$7.8147\times10^{+1}$	$-3.3247\times10^{+0}$	3.0088×10^{-2}	
61	61	60	$-3.9588\times10^{+2}$	$5.3928\times10^{+1}$	$-1.8871\times10^{+0}$	1.0722×10^{-2}		
61	65	66	$-1.5241\times10^{+3}$	$2.0762\times10^{+2}$	$-7.2652\times10^{+0}$	4.1280×10^{-2}		
62	62	20	$-2.0643\times10^{+3}$	$2.2298\times10^{+3}$	$-8.2955\times10^{+2}$	$1.3217\times10^{+2}$	$-8.7379\times10^{+0}$	1.7050×10^{-1}
62	62	40	$4.9195\times10^{+2}$	$-1.7981\times10^{+2}$	$2.1490\times10^{+1}$	-9.1429×10^{-1}	8.2741×10^{-3}	
62	62	60	$6.9674\times10^{+2}$	$-9.4914\times10^{+1}$	$3.3212\times10^{+0}$	-1.8871×10^{-2}		
64	62	66	$1.2193\times10^{+4}$	$-1.6610\times10^{+3}$	$5.8122\times10^{+1}$	-3.3024×10^{-1}		
64	64	20	$3.4405\times10^{+2}$	$-3.7163\times10^{+2}$	$1.3826\times10^{+2}$	$-2.2028\times10^{+1}$	$1.4563\times10^{+0}$	-2.8416×10^{-2}
64	64	40	$-3.5778\times10^{+3}$	$1.3077\times10^{+3}$	$-1.5629\times10^{+2}$	$6.6494\times10^{+0}$	-6.0175×10^{-2}	
64	64	60	$2.1113\times10^{+2}$	$-2.8762\times10^{+1}$	$1.0064\times10^{+0}$	-5.7184×10^{-3}		
65	61	66	$-1.5241\times10^{+3}$	$2.0762\times10^{+2}$	$-7.2652\times10^{+0}$	4.1280×10^{-2}		
65	65	20	$8.6012\times10^{+1}$	$-9.2907\times10^{+1}$	$3.4564\times10^{+1}$	$-5.5069\times10^{+0}$	3.6408×10^{-1}	-7.1040×10^{-3}
65	65	40	$-1.1181\times10^{+2}$	$4.0865\times10^{+1}$	$-4.8842\times10^{+0}$	2.0779×10^{-1}	-1.8805×10^{-3}	
65	65	60	$-6.5979\times10^{+1}$	$8.9880\times10^{+0}$	-3.1451×10^{-1}	1.7870×10^{-3}		
66	66	20	$2.0643\times10^{+3}$	$-2.2298\times10^{+3}$	$8.2955\times10^{+2}$	$-1.3217\times10^{+2}$	$8.7379\times10^{+0}$	-1.7050×10^{-1}
66	66	40	$2.0125\times10^{+3}$	$-7.3557\times10^{+2}$	$8.7915\times10^{+1}$	$-3.7403\times10^{+0}$	3.3849×10^{-2}	
66	66	60	$3.1670\times10^{+2}$	$-4.3143\times10^{+1}$	$1.5097\times10^{+0}$	-8.5776×10^{-3}		

nates (θ, ϕ), the lattice will develop a corresponding strain. This is the flexible-lattice model. The two models give different contributions to the macroscopic-anisotropy constants. The two models have been discussed in connection with spin-wave and resonance experiments by Cooper.[3]

In equilibrium with respect to strains, the combined contribution of the elastic and the magnetoelastic terms in the Hamiltonian is minus the elastic term (8). The contribution from the irreducible strains (6) to the anisotropy constants is therefore in the frozen- and flexible-lattice models obtained from

$$\frac{\partial}{\partial\theta}F(\theta,\phi)=\left\langle\frac{\partial}{\partial\theta}\left(-\frac{1}{2}\sum_{\substack{\Gamma\\ ij}}c_{ij}^{\Gamma}\epsilon^{\Gamma,i}\epsilon^{\Gamma,j}\right)\right\rangle$$

flexible-lattice model

$$=-\sum_{\substack{\Gamma\\ ij}}c_{ij}^{\Gamma}\langle\epsilon^{\Gamma,i}\rangle\left\langle\frac{\partial}{\partial\theta}\epsilon^{\Gamma,j}\right\rangle,$$

frozen-lattice model. (17)

$\partial F(\theta,\phi)/\partial\phi$ is obtained analogously. Under the average sign $\langle\dots\rangle$ the Stevens-operator expressions for the strains (7) are to be used.

In the frozen-lattice model the effect is simply to alter the single-ion anisotropy contribution B_L^M in (14) and (16):

$$B_L^M\to B_L^M-(B_{LM}^{\alpha,1}\langle\epsilon^{\alpha,1}\rangle+B_{LM}^{\alpha,2}\langle\epsilon^{\alpha,2}\rangle)\,,\quad M=0,6\,. \tag{18}$$

Since the frozen strains just add a contribution to the crystalline field, only strains with hexagonal symmetry contribute. Two-ion anisotropy contributes in the molecular field theory by symmetry in a similar fashion by

$$-\frac{2}{N}\sum_{\substack{l'm'i'\\ f,g}}K_{LMc}^{l'm'i'}(f,g)\langle O_{l'}^{m'}(i')\rangle\,. \tag{19}$$

In the flexible-lattice model we again, for simplicity, limit ourselves to terms which transform according to the hexagonal point group. Therefore we need only consider terms in $\epsilon^{\Gamma,i}\epsilon^{\Gamma,j}$ which have L even and $M=0$ or $M=6$ when transformed to products of Stevens operators and reduced to one Stevens operator O_L^M. We find for the α, γ, and ϵ strains

$$\frac{1}{2}\sum_{i,j}c_{ij}^{\alpha}\epsilon^{\alpha,i}\epsilon^{\alpha,j}=\frac{1}{2}\sum_{\substack{i\neq j\\ l'm'\\ l''m''}}\frac{1}{c_{ii}^{\alpha}c_{jj}^{\alpha}-(c_{ij}^{\alpha})^2}$$

$$\times(c_{jj}^{\alpha}B_{l'm'}^{\alpha,i}B_{l''m''}^{\alpha,i}-c_{ij}^{\alpha}B_{l'm'}^{\alpha,i}B_{l''m''}^{\alpha,j})O_{l'}^{m'}(c)O_{l''}^{m''}(c)\,,$$

$$\frac{1}{2}\sum_{i}c^{\Gamma}(\epsilon^{\Gamma,i})^2=\frac{1}{c^{\Gamma}}\frac{1}{2}\sum_{\substack{l'm'\\ l''\\ p=(c,s)=(1,2)}}\Bigg[B_{l'm'}^{\Gamma}B_{l''m'}^{\Gamma}$$

$$\times O_{l'}^{m'}(p)O_{l''}^{m'}(p)$$

$$-(-1)^p\sum_{m''\neq m'}B_{l'm'}^{\Gamma}B_{l''m''}^{\Gamma}O_{l'}^{m'}(p)O_{l''}^{m''}(p)\Bigg], \tag{20}$$

where m' and m'' can take the values according to (7). The L, M term in the reduction of the pair of Stevens operators is[6]

$$O_{l'}^{m'}(c)O_{l''}^{m''}(c)\Big|_L^M=[l'm',l''m'',LM]\,O_L^M(c)\,.$$

The relevant coefficients $[l'm',l''m'',LM]$ are given in Table I. $[l'm',l''m'',LM]=[l''m'',l'm',LM]$ when $l'+l''+L$ is even. The reduction of the relevant sine-type Stevens operators can be obtained from the following identity:

$$O_{l'}^{m'}(c)O_{l''}^{m''}(c)\pm O_{l'}^{m'}(s)O_{l''}^{m''}(s)\Big|_l^{m'\pm m''}=0\quad(m',m''\neq0)\,.$$

The result of the magnetoelastic coupling in the flexible-lattice model is, therefore, that the single-ion anisotropy contribution B_L^0 [(14) and (16)] is replaced by

$$B_L^0\to B_L^0-\sum_{\substack{m\\ l'l''\\ i\neq j}}\frac{1}{c_{ii}^{\alpha}c_{jj}^{\alpha}-(c_{ij}^{\alpha})^2}$$

$$\times(c_{jj}^{\alpha}B_{l'm}^{\alpha,i}B_{l''m}^{\alpha,i}-c_{ij}^{\alpha}B_{l'm}^{\alpha,i}B_{l''m}^{\alpha,j})[l'm,l''m,L0]$$

$$-\sum_{\substack{l'l''\\ m}}\left(\frac{1}{c^{\gamma}}B_{l'm}^{\gamma}B_{l''m}^{\gamma}+\frac{1}{c^{\epsilon}}B_{l'm}^{\epsilon}B_{l''m}^{\epsilon}\right)$$

$$\times[l'm,l''m,L0]\,. \tag{21}$$

There is only a contribution to the B_L^0 term from the $m'=m''$ terms. The magnetoelastic contribution to the sixfold anisotropy comes from the $m'\neq m''$ terms

$$B_6^6\to B_6^6-\frac{1}{2}\sum_{\substack{l'l''\\ m'\neq m''}}\left(\frac{1}{c^{\gamma}}B_{l'm'}^{\gamma}B_{l''m''}^{\gamma}\right.$$

$$\left.+\frac{1}{c^{\epsilon}}B_{l'm'}^{\epsilon}B_{l''m''}^{\epsilon}\right)[l'm',l''m'',66]\,. \tag{22}$$

Equations (21) and (22) show the magnetoelastic contribution to the anisotropy when the lattice is strained according to the average moment direction. Its temperature dependence is related to the rapid moment precession and is likely to be well approximated by the frozen-lattice model. In this case the temperature dependence follows that corresponding to l' and l'' in (20) and not that of L in (21) and (22).

The large number of parameters in the above expressions can then be reduced considerably. By measuring the equilibrium strains in thermodynamic equilibrium, (7), it is possible to determine certain combinations of the parameters B_{lm}^{j}. In particular,[3] the magnetostriction coefficients $\mathfrak{C}$ and $\mathfrak{A}$ are the coefficients of the γ strain, which transform under rotation around the c axis as $\cos2\phi$ and $\cos4\phi$, respectively:

$$\frac{1}{c^{\gamma}}\sum_{l}B_{l2}^{\gamma}\langle O_l^2(c)\rangle=\mathfrak{C}\cos2\phi\,,$$

$$\frac{1}{c^{\gamma}}\sum_{l}B_{l4}^{\gamma}\langle O_l^4(c)\rangle=-\tfrac{1}{2}\mathfrak{A}\cos4\phi\,. \tag{23}$$

Notice that the Stevens operators are in a c-axis representation. Magnetostriction coefficients for the ϵ strain, $\mathfrak{D}$ and $\mathfrak{B}$, transforming as $\cos\phi$ and $\cos 5\phi$, respectively, may be defined analogously according to (7). The contribution in the frozen-lattice model for the strained crystal is

$$B_6^6 \to B_{6\,\text{single ion}}^6 + \tfrac{1}{2}c^{\gamma}\mathfrak{A}\mathfrak{C} + \tfrac{1}{2}c^{\epsilon}\mathfrak{B}\mathfrak{D} \,,$$

and for the axial anisotropy it is $c^{\gamma}(\mathfrak{C}^2 + \frac{1}{4}\mathfrak{A}^2) + c^{\epsilon}(\mathfrak{D}^2 + \frac{1}{4}\mathfrak{B}^2)$. The macroscopic coefficients do not adequately describe the contribution for each value of L. However, it can be found under simplifying assumptions by means of (20), (21), and Table I. The ϵ strain only contributes if the magnetization has a component along the c axis.

IV. SPIN-WAVE THEORY OF TEMPERATURE DEPENDENCE OF RESONANCE FREQUENCY AND STEVENS OPERATORS

In the limit where the isotropic exchange interaction is the dominant term in the Hamiltonian, the temperature dependence of the resonance frequency and the Stevens operators can be calculated in a renormalized spin-wave theory. Bose-operator expansions of the Stevens operators have been calculated by Lindgård and Danielsen[6] using a method where the Stevens operators are formally expanded in a well-ordered Bose-operator series, the coefficients of which are found by matching the matrix elements in and between the ground state and the excited states. This method is simpler than, but equivalent to, the Holstein-Primakoff transformation. We find the temperature dependence of the Stevens operator $O_l^m(c)$ by

$$\langle O_l^m(c)\rangle = \mathrm{Tr}[O_l^m(c)e^{-H/k_BT}]/\mathrm{Tr}(e^{-H/k_BT}) \,. \qquad (24)$$

The operators are expanded in spin-wave operators and the trace is taken over the spin-wave states. We include consistently two-magnon interaction terms (four Bose-operator terms) and decouple these by means of the Hartree-Fock approximation.

A. Spin-wave theory in the Hartree-Fock approximation: Temperature dependence of the resonance frequency

Let us choose a coordinate system in which the z direction is along the quantization axis, i.e., the average moment direction. The Hamiltonian then contains no sine-type Stevens operators and has the form

$$H = H_{\mathrm{ex}} + \sum_{\substack{l,m\\ i}} K_l^m O_l^m(c)_i \,, \quad m = 0, 2, 4, 6\,; \ \text{all } l, \qquad (25)$$

where the coefficients K_l^m are obtained by properly rotating the contributions from the single-ion anisotropy and magnetostrictive terms and two-ion anisotropy (19). According to Lindgård and Danielsen[6] the Stevens operators belonging to the site i are expanded in terms of Bose operators as follows:

$$O_l^0 = C(l,0)S_l\Big[1 - \alpha_l\frac{1}{S_1}a^{\dagger}a + \alpha_l(\alpha_l-1)/4\,\frac{1}{S_2}a^{\dagger}a^{\dagger}aa + \cdots\Big] \,,$$

$$O_l^2 = C(l,2)\frac{S_l}{(S_2)^{1/2}}\frac{1}{2}\Big[(aa + a^{\dagger}a^{\dagger}) - \Big(\frac{S_1S_2}{S_3}\Big)^{1/2} \times\Big(\frac{\alpha_l}{3} - 1 + \Big(\frac{S_1S_3}{S_2}\Big)^{1/2} - \frac{S_3}{S_2}\Big)\frac{1}{S_1}(a^{\dagger}a^{\dagger}a^{\dagger}a + a^{\dagger}aaa) + \cdots\Big] \qquad (26)$$

$$O_l^4 = C(l,4)\frac{S_l}{(S_4)^{1/2}}[(a^{\dagger}a^{\dagger}a^{\dagger}a^{\dagger} + aaaa) + \cdots\ ] \,,$$

where $\alpha_l = \frac{1}{2}l(l+1)$,

$S_l = J(J-\frac{1}{2})(J-1)\cdots[J-\frac{1}{2}(l-1)]$, $l = 1, 2, 3, \ldots,$

and the coefficients $C(l, m)$ are given in Table II for l even only.

The interaction terms in the Hartree-Fock approximation are, in the direct space,

$$\begin{aligned}
&[(1/J^2)a^{\dagger}a^{\dagger}aa\,]_{\mathrm{HF}} \\
&\quad = (1/2J)(aa + a^{\dagger}a^{\dagger})2b(T) - b(T)^2 \\
&\qquad + (1/J)a^{\dagger}a4\Delta M(T) - 2\Delta M(T)^2 \,, \\
&(1/J^2)(a^{\dagger}a^{\dagger}a^{\dagger}a + a^{\dagger}aaa)_{\mathrm{HF}} \\
&\quad = (1/2J)(aa + a^{\dagger}a^{\dagger})6\Delta M(T) \\
&\qquad + (1/J)a^{\dagger}a6b(T) - 6\Delta M(T)b(T) \,, \\
&(1/J^2)(a^{\dagger}a^{\dagger}a^{\dagger}a^{\dagger} + aaaa)_{\mathrm{HF}} \\
&\quad = (1/2J)(aa + a^{\dagger}a^{\dagger})12b(T) - 6b^2(T) \,,
\end{aligned} \qquad (27)$$

where the characteristic functions are defined as

$$\Delta M(T) = (1/J)\,\langle a^{\dagger}a\rangle$$

and

$$b(T) = (1/J)\langle aa\rangle = (1/J)\langle a^{\dagger}a^{\dagger}\rangle \,,$$

where the Bose operators act on a single site i. The characteristic function $\Delta M(T)$ is related to the temperature-dependent deviation of the reduced magnetization $m(T)$ by

$$\langle J_z\rangle = \langle O_1^0(c)\rangle = J[1 - \Delta M(T)] = J[1 - \Delta M(0)]m(T) \,. \qquad (28)$$

The characteristic function $b(T)$ is related to the nonspherical precession of the angular momentum in the presence of anisotropy,

$$\langle J_x^2\rangle - \langle J_y^2\rangle = \langle O_2^2(c)\rangle = 2J^2b(T)[1 - \tfrac{3}{2}\Delta M(T)] \,. \qquad (29)$$

The effective Hamiltonian for the noninteracting Bose operators is then, after the usual Fourier transformation to wave-vector space as given by Lindgård *et al.*,[4]

$$H = \frac{1}{N}\sum_q\,[A_q(T)\tfrac{1}{2}(a_q^{\dagger}a_q + a_qa_q^{\dagger}) + B_q(T)\tfrac{1}{2}(a_{-q}a_q + a_q^{\dagger}a_{-q}^{\dagger})] \,. \qquad (30)$$

A diagonalization gives the spin-wave energy

$$E_q(T) = \{[A_q(T) - B_q(T)][A_q(T) + B_q(T)]\}^{1/2} . \qquad (31)$$

The transformation which diagonalizes the Hamiltonian enables us to evaluate the characteristic functions in terms of $E_q(T)$, $A_q(T)$, and $B_q(T)$:

$$\begin{aligned} \Delta M(T) &= \frac{1}{JN} \sum_q \langle a_q^\dagger a_q \rangle \\ &= \frac{1}{JN} \sum_q \left(\frac{A_q(T)}{E_q(T)} [n_q(T) + \tfrac{1}{2}] - \frac{1}{2} \right) , \\ b(T) &= \frac{1}{JN} \sum_q \langle a_{-q} a_q \rangle \\ &= \frac{-1}{JN} \sum_q \frac{B_q(T)}{E_q(T)} [n_q(T) + \tfrac{1}{2}] , \end{aligned} \qquad (32)$$

where $n_q(T) = 1/(e^{E_q(T)/k_B T} - 1)$ is the spin-wave population factor and the summation is over N points in the Brillouin zone corresponding to the number of atoms in the crystal. We shall here only treat the resonance frequency, i.e., the spin-wave energy gap $E_{q=0}$. In this case the contribution from the exchange interaction vanishes, and the number of atoms per unit cell is irrelevant. Using (25) and (27) we find, to the first order in the characteristic functions $\Delta M(T)$, $b(T)$, and $1/J$, the following contributions to the temperature dependence of the energy gap, valid for all l:

$$\begin{aligned} [A_0(T) \pm B_0(T)]^{\mathrm{HF}}_{\mathrm{SW}} = \frac{1}{J} \sum_l S_l (&- K_l^0 C(l, 0) \\ &\times \alpha_l \{1 - (\alpha_l - 1)(1 + 1/2J)[\Delta M(T) \pm \tfrac{1}{2} b(T)]\} \\ &\pm K_l^2 C(l, 2)(1 + 1/4J)\{1 - (\alpha_l - 1)(1 + 1/2J) \\ &\times [\Delta M(T) \pm \tfrac{1}{2} b(T)]\}) + \delta^\pm(T) , \end{aligned} \qquad (33)$$

where

$$\begin{aligned} \delta^\pm(T) = \frac{1}{J} \sum_l S_l \tfrac{1}{2}(1 + 1/2J) & \\ \times (\{ - (1 + 1/4J) K_l^2 C(l, 2)(\alpha_l - 1) &+ (1 + 1/J) \\ \times [K_l^2 C(l, 2) \pm 24 K_l^4 C(l, 4)]\} b(T) & \\ \pm (1 + 1/J) K_l^2 C(l, 2) \Delta M(T)) . & \end{aligned} \qquad (34)$$

$\delta^\pm$ is zero in the case of pure axial anisotropy, i.e., when $K_l^{2,4} = 0$ for all l. For the case of pure planar anisotropy the resonance energy is zero for all temperatures, according to the Goldstone theorem; consequently $\delta^\pm$ must be zero and the factor $(1 + 1/4J)$ in (33) is unphysical. The problem arises because of the approximate Hartree-Fock treatment of the dynamical interactions. We shall discuss this in a forthcoming paper[19] and here neglect $\delta^\pm$ and the $(1 + 1/4J)$ factor. The planar anisotropy case is treated in more detail in the Appendix.

Cooper[3] has discussed the temperature dependence of the energy gap on the basis of a classical formula for the resonance frequency[10] and found the following contribution to the temperature dependence:

$$\begin{aligned} [A_0(T) \pm B_0(T)]_{\mathrm{class}} & \\ = \frac{1}{J} \sum_l J^l \{ & - K_l^0 C(l, 0) \alpha_l \hat{I}_{(2l+1)/2} \\ & \times [\mathcal{L}^{-1}(m(T))]/m(T) \pm K_l^2 C(l, 2) \hat{I}_{(2l+1)/2} \\ & \times [\mathcal{L}^{-1}(m(T))]/m(T) \} . \end{aligned} \qquad (35)$$

Here $\hat{I}_{(2l+1)/2}[\mathcal{L}^{-1}(m(T))]$ is the ratio of the hyperbolic Bessel function of the order of $(2l + 1)/2$ to that of the order of $\frac{1}{2}$, where the argument of the Bessel function is the inverse Langevin function of the reduced magnetization. When the reduced magnetization is close to 1, we can expand in the deviation $\Delta M(T)$ as follows:

$$\begin{aligned} \hat{I}_{(2l+1)/2}[\mathcal{L}^{-1}(m(T))]/m(T) &\sim m(T)^{\alpha_l - 1} \\ &\sim [1 - (\alpha_l - 1)\Delta M(T)] . \end{aligned}$$

Thus if we neglect the ellipticity parameter $b(T)$ in (33), we obtain in the Hartree-Fock approximation, and for $J \gg 1$, exactly Cooper's result. However, the inclusion of the ellipticity parameter $b(T)$ significantly alters this result. We shall see later for Tb and Dy that $\Delta M(T)$ and $b(T)$ are of similar magnitude. Therefore the constants K_l^m cannot be scaled equally in the brackets containing $A + B$ and $A - B$ as suggested by the expression of Cooper.

However, the simplicity of Cooper's result is retained. According to (33) it is possible to find the temperature renormalization of the energy gap by scaling each term of order l appropriately. The numerous works,[3,5,18] which in more or less detail discuss the anisotropy and magnetoelastic contributions, can thus directly be renormalized by means of (33).

B. Temperature dependence of the Stevens operators

Using the Bose expansion for the Stevens operators (26) and taking the trace over spin-wave states according to (24), we find for the temperature dependence of the average value of the Stevens operators to the second order in $\Delta M(T)$ and $b(T)$ and the

TABLE II. Coefficients in the expressions (26), (33), and (36).

l	$C(l, 0)$	$C(l, 2)$	$C(l, 4)$
1	1		
2	2	2	
3	2	2	
4	8	12	2
5	8	4	2
6	16	32	20

first order in $1/J$,

$$\begin{aligned}\langle O_l^0(c)\rangle_T &= S_l C(l,0)\{1-\alpha_l \Delta M(T)\\ &\quad+\tfrac{1}{2}\alpha_l(\alpha_l-1)(1+1/2J)[\Delta M(T)^2+\tfrac{1}{2}b(T)^2]+\cdots\},\\ \langle O_l^2(c)\rangle_T &= S_l C(l,2)(1+1/4J)b(T)\\ &\quad\times\{1-[\alpha_l-1-\tfrac{1}{2}(1+3/4J)](1+1/2J)\Delta M(T)+\cdots\},\\ \langle O_l^4(c)\rangle_T &= S_l C(l,4)(1+3/2J)6b(T)^2.\end{aligned}\tag{36}$$

In the large angular-momentum limit we can write the temperature dependence of the Stevens operators as power laws of the reduced magnetization and terms involving the ellipticity parameter $b(T)$. For a small deviation from saturation we can expand the reduced magnetization to a power in $\Delta M(T)$,

$$\begin{aligned}m(T)^\alpha = [1-\Delta M(T)]^\alpha \sim 1 - \alpha\Delta M(T)\\ +\tfrac{1}{2}\alpha(\alpha-1)\Delta M(T)^2+\cdots.\end{aligned}\tag{37}$$

From (36) we then find the following power laws:

$$\begin{aligned}\langle O_l^0(c)\rangle_T &= \langle O_l^0(c)\rangle_0 m(T)^{\alpha_l}[1+b(T)^2]^{\alpha_l(\alpha_l-1)/4},\\ \langle O_l^2(c)\rangle_T &= \langle O_l^2(c)\rangle_0 m(T)^{\alpha_l-3/2}b(T),\\ \langle O_l^4(c)\rangle_T &= \langle O_l^4(c)\rangle_0 b(T)^2,\end{aligned}\tag{38}$$

which are valid for large angular momentum J and at low temperatures. The identification of power laws from the finite expansions (36) is of course not unique. However, we have shown that the renormalized spin-wave theory in the Hartree-Fock approximation gives the first two terms of the $\alpha_l = \frac{1}{2}l(l+1)$ power law exactly. When the ellipticity parameter $b(T)$ can be neglected, we recover the well-known $\frac{1}{2}l(l+1)$ power law, which is discussed in the work by Callen and Callen.[2] The correction to this law due to the nonspherical spin precession has recently been discussed by Brooks[5] and by Brooks and Egami.[16] However, their results differ from ours since they did not treat the kinematic effects systematically when decoupling the equation of motions for the angular-momentum operators.

The temperature dependence of the strains and of the macroscopic-anisotropy constants derived in the previous section can be found by inserting the results for the temperature dependence of the Stevens operators.

C. Relation between the macroscopic and microscopic anisotropy parameters in the spin-wave theory

Within the framework of the spin-wave theory and the Hartree-Fock (HF) approximation we can evaluate the relation between the macroscopic and microscopic anisotropy parameters directly. Let us consider an anisotropic magnet in a magnetic field which tilts the direction of magnetization to a position described by the polar coordinates (θ, ϕ). The direction in zero field is (θ_0, ϕ_0). In the HF spin-wave theory the Hamiltonian for this system takes the form

$$H(\theta,\phi) = A + a(\theta,\phi) + \sum_q [E_q + \epsilon_q(\theta,\phi)]\hat{n}_q, \tag{39}$$

where A and $a(\theta, \phi)$ are constants independent of and dependent on the angles (θ, ϕ), respectively. E_q and $\epsilon_q(\theta, \phi)$ are, respectively, the parts of the renormalized spin-wave energy which are independent of and dependent on, the angles. By definition $a(\theta_0, \phi_0) = \epsilon_q(\theta_0, \phi_0) = 0$. The spin-wave number operator is $\hat{n}_q$, and the summation is over the Brillouin zone. We likewise separate the free energy in a part F independent of (θ, ϕ) and a part $f(\theta, \phi)$ which depends on the angles. According to (10) we have

$$\begin{aligned}e^{-\beta[F+f(\theta,\phi)]} = e^{-\beta[A+a(\theta,\phi)]}\\ \times\sum_{n_q'=0}^{\infty}\left(e^{-\beta}\sum_q[E_q+\epsilon_q(\theta,\phi)]n_q'\right),\end{aligned}$$

which gives

$$\begin{aligned}F+f(\theta,\phi) = A + a(\theta,\phi) - k_B T\\ \times\sum_q \ln\frac{1}{1-e^{-\beta[E_q+\epsilon_q(\theta,\phi)]}}.\end{aligned}\tag{40}$$

Assuming that the system is dominated by the isotropic exchange interaction, we can expand to first order in the angular-dependent terms. The part of the free energy $f(\theta, \phi)$ which depends on the angles is then, to first order in $\epsilon(\theta, \phi)$,

$$f(\theta,\phi) = a(\theta,\phi) + \sum_q \epsilon_q(\theta,\phi)n_q^0, \tag{41}$$

where $n_q^0 = 1/(e^{\beta E_q} - 1)$ is the occupation number when the magnetization is along the (θ_0, ϕ_0) direction.

The result (41) is identical with the angular-dependent part of the internal energy evaluated as the zero-field thermal average of the angular-dependent terms in (39). The free energy obtained by equating the angular derivatives (12) and (13) and evaluating the averages in the HF spin-wave theory (24) gives the identical result, i.e., the internal energy. We conclude that the approximation involved in the expressions for the anisotropy constants $K_n(T)$ in (14) and $\kappa_l(T)$ in (16) is valid at low temperatures and in the limit where the anisotropy is much smaller than the isotropic interaction. Only in this limit is the assumed angular dependence of the free energy (11) correct for all (θ, ϕ). At high temperature and when the anisotropy is large (still small enough to be treated by spin-wave theory), the free energy $f(\theta, \phi)$ associated with the magnetization in an arbitrary direction (θ, ϕ) must be evaluated directly from (40), including the effect of the magnetic field which is necessary to tilt the magnetization. The free energy thus becomes field dependent. However, a differential measurement which only tilts the magnetization infinitesimally from the equilibrium direction (θ_0, ϕ_0) can still be interpreted on the basis of (14) and (16). Care must be taken in this case as the contribution from hexagonal terms cannot be separated from that from the nonhexagonal terms in a distorted crystal.

It should finally be mentioned that the slightly different theoretical approach by Brooks and Egami[16] is subject to precisely the same limits of validity as mentioned above. The main restriction in their theory and in the present spin-wave theory is that the anisotropy is assumed not to perturb the ground state significantly from a pure $|J\rangle$ state. If the anisotropy has a magnitude and nature which makes this assumption invalid a different approach to the problem is necessary.

V. DISCUSSION AND NUMERICAL RESULTS

Let us apply the theory to the following three ferromagnetic rare-earth metals: gadolinium, terbium, and dysprosium. Gd^{3+} is an S-state ion $(S=\frac{7}{2})$ and gadolinium consequently has a negligible anisotropy in comparison with terbium and dysprosium, in which the large orbital angular momentum $(L=3, 5,\ S=3, \frac{5}{2}$, and $J=6, \frac{15}{2}$, respectively) causes a large anisotropy, which confines the total angular momentum J to a particular direction in the basal plane. However, the exchange interaction is dominant and the excitations in all materials can be treated by the spin-wave theory. The measured spin-wave dispersion curves for Gd (Ref. 11) show (i) no energy gap, as we would expect, and (ii) that the interaction is significantly stronger in the basal planes than between the planes. The measured dispersion curves for Tb (Ref. 12) and Dy (Ref. 13) are similar, but with a substantial energy gap due to the anisotropy.

On the basis of the measured dispersion curves E_q and (32) we can evaluate the temperature dependence of the magnetization and the ellipticity parameter $b(T)$ by summation over the Brillouin zone. It is necessary to sum numerically over the total zone. In the calculation we used the following procedure to obtain the spin-wave energies at all wave vectors q. The spin-wave energy has for the acoustic branch the form $E_q^2=A_q^2-B^2$, where B to a good approximation is independent of q since the anisotropy is dominantly of single-ion origin. To obtain accurate results for $\Delta M(T)$ and $b(T)$ at low temperatures, A_q^2 is fitted to the polynominal expansion

$$A_q^2=A_0^2+\sum_{n=1}^{4}\left(c_\perp^{(n)}q_\perp^{2n}+c_\parallel^{(n)}q_\parallel^{2n}\right)$$

for $q_\perp$ in the basal plane less than $0.1q_{\perp\mathrm{zone}}$ and $q_\parallel$ along the c axis less than $0.4q_{\parallel\mathrm{zone}}$. This analytic form of E_q agrees with the measured dispersion curves within the experimental accuracy for energies below 5 meV (~60 K). The detailed shape of the spin-wave dispersion surface at higher energies is of less importance in the calculation of $\Delta M(T)$ and $b(T)$. The surface for this part of the zone is approximated by linear expressions in $q_\perp$ and $q_\parallel$. The optic spin-wave branch is approximated by a constant. The data are summarized in Table III together with the calculated values of $\Delta M(0)$ and $b(0)$. We note that the zero-point motion caused by the anisotropy gives a reduction of the saturation moment for the fully aligned ferromagnetic ground state. For Tb and Dy it amounts to 0.59% and 0.76%—corresponding to $0.05\mu_B$ and $0.08\mu_B$, respectively. This correction must be included when the polarization of the conduction electrons is estimated from the measured saturation magnetic moments. We thus find the values given in Table III.

It is well known from the extensive studies of the antiferromagnetic ground-state problem[14] that spin-wave theory is unsatisfactory in the study of the ground state. Therefore the calculated zero-point deviation $\Delta M(0)$ must be taken with some reservation. The same applies to the ellipticity parameter $b(0)$, which for Tb and Dy is quite large, 0.031 and 0.032, respectively.

The temperature dependence of $\Delta M(T)$ and $b(T)$ caused by the population of the excited states is expected to be accurately given by the spin-wave theory. The results are shown in Fig. 1. We note

TABLE III. Spin-wave dispersion data for Gd, Tb, and Dy in units of meV. A_0 and B_0 are defined in the text (33), (A5). Δ is the energy gap. Shown also are the values used for the zone-boundary energies E_q^{zb} and the optic energy. Summation over the Brillouin zone of (32) gives the $T=0$ values for $\Delta M(T)$ and $b(T)$. Given also is the conduction-electron polarization $M_{\mathrm{c.e.}}$, derived from the magnetization data [Ref. 15(a)] on the basis of the present theory.

	A_0	$-B_0$	Δ	$E_{q\parallel}^{\mathrm{zb}}$	$E_{q\perp}^{\mathrm{zb}}$	$\bar{E}_{\mathrm{optic}}$	$\Delta M(0)$	$b(0)$	$M_{\mathrm{c.e.}}\mu_B$
Gd			0	10.0	15.0	15.0	0	0	0.55
Tb	4.25	3.90	1.69	8.0	12.0	12.0	0.0059	0.0309	0.41
Dy	4.82	3.60	3.21	6.0	8.0	8.0	0.0076	0.0324	0.41

the interesting fact that because of the two-dimensional character of the dispersion relation for Gd, the temperature dependence of the relative magnetization $m(T)$ is much greater than that of a Brillouin function for $S=\frac{7}{2}$. In fact, $m(T)$ follows closely that of Tb, although Tb has a large energy gap. This is in agreement with experiments,[15] as shown in Fig. 1. The temperature dependence of $m(T)$ for Dy is greater on account of the weaker exchange interaction, in agreement with experiments.[15] The temperature dependence of the ellipticity parameter $\delta b(T) = b(T) - b(0)$ is also shown in Fig. 1, right scale. The reason why $b(0)$ is larger than $\Delta M(0)$ is clear from (32), which shows that $\Delta M(0)$ involves a difference of almost equal terms. The temperature dependence of $\Delta M(T)$ is greater than that of $b(T)$ because A_q is an increasing function of q; therefore the contributions to $\Delta M(T)$ are large when the states at higher energies (and larger wave vectors) become populated with increasing temperature. We note that the ratio $[b(T)-b(0)]/[\Delta M(T)-\Delta M(0)]$ is approximately $\frac{1}{3}$ over the whole temperature region for Tb and Dy. This means that the ellipticity corrections of the temperature laws for the Stevens operators (36) and, in particular, for the spin-wave energy gap [(31) and (33)] are very significant for Tb and Dy. The terms of the order of l in $[A_0(T) \pm B_0(T)]$, the curly brackets in Eq. (33), have the approximate temperature dependence proportional to $C_l^{\pm} m(T)^{\beta_l^{\pm}}$, where the corrected exponent is

$$\beta_l^{\pm} = (\alpha_l - 1)(1 + 1/2J)[1 - \Delta M(0)](1 \pm \tfrac{1}{6})/C_l^{\pm} ,$$

and the scale factor is

$$C_l^{\pm} = 1 - (\alpha_l - 1)(1 + 1/2J)[\Delta M(0) \pm \tfrac{1}{2} b(0)] .$$

We note that the corrected exponents $\beta_l^{\pm}$ differ significantly from the classical exponents $\alpha_l - 1$. This must be taken into account if the temperature dependence is used to extract information about the relative magnitude of terms with different order l.

TABLE IV. Exponents and scale factors for the resonance energy and the macroscopic anisotropy in the classical theory and in the present theory for Tb and Dy. For the dominant magnetoelastic contribution (γ strain) the exponents are discussed in the Appendix. For odd values of l, K_l^m in (25) must also contain average values of Stevens operators with odd l values, for which the macro scopic exponents should be used. The $l=6$ results are in particular sensitive to uncertainties in $\Delta M(0)$ and $b(0)$.

	Resonance energy				Macroscopic anisotropy			
l	$\alpha_l - 1$	β_l^+	β_l^-	C_l^+	C_l^-	α_l	β_l^M	C_l^M
1	0	0	0	1	1	1	1	0.99
2	2	2.6	1.8	0.95	1.02	3	2.6	1.02
3	5	7	4.3	0.88	1.05	6	4.7	1.06
4	9	14	7.4	0.79	1.09	10	7.0	1.13
5	14	26	11	0.68	1.14	15	9.5	1.22
6	20	(46)	(15)	(0.54)	(1.21)	21	(12)	(1.35)

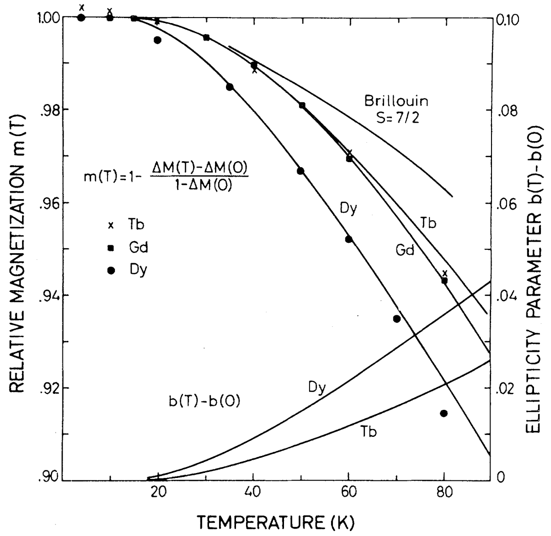

FIG. 1. Relative magnetization $m(T)$ and temperature variation of the ellipticity parameter $b(T)$ for Gd, Tb, and Dy. Experimental points are taken from Ref. 15.

However, the calculation shows that it is valid to use $m(T)$ in a parametrization of the temperature dependence. The scale factors are of importance because $C_l^+ \neq C_l^-$ and are different from those to be used for the macroscopic anisotropy parameters.

The temperature dependence of magnetoelastic terms in the frozen-lattice model involves both the exponents $\beta_l^{\pm}$ and exponents β_l^{ϵ} relevant for the strains. The temperature dependence of the strains can be obtained by rotating the Stevens operators in (7) and using (36). The magnetoelastic effect when the moment is in the basal plane is discussed in the Appendix.

The temperature dependence of the anisotropy parameters which can be measured by macroscopic measurements is most easily found from the expressions for the anisotropy coefficients $\kappa_l(T)$, Eq. (16). When we use the temperature laws for the Stevens operators (36), we find that $\kappa_l(T)$ is proportional to

$$1 - [\tfrac{1}{2} l(l+1)]\Delta M(T) + [\tfrac{1}{2} l(l-1)](1 + 1/4J) b(T) + \cdots ,$$

which can for Tb and Dy approximately be written as $C_l^M m(T)^{\beta_l^M}$, where the macroscopic exponent is

$$\beta_l^M = [\alpha_l - \tfrac{1}{3}\alpha_{l-1}(1 + 1/4J)]/C_l^M ,$$

and the macroscopic scale factor is

$$C_l^M = 1 - \alpha_l \Delta M(0) + \alpha_{l-1}(1 + 1/4J) b(0) .$$

The numerical values are given in Table IV. The scale factors for the macroscopic measurements C_l^M are thus different from those relevant for the microscopic measurements $C_l^{\pm}$. This may account for the difference in magnitude of the anisotropy parameters previously determined[1] without taking this difference into account. Spin-wave measurements have not so far been analyzed on the basis of the spin-wave theory in the Hartree-Fock approximation [(31) and (33)]. The correction of the temperature exponents for the macroscopic-anisotropy coefficients is particularly large for the $l=6$ term. This is in agreement with experiments[15] which show large deviations from the classical $\frac{1}{2}l(l+1)$ law for the order of $l=6$. The magnetoelastic contributions are also important in this case, (23) and (A4).

VI. CONCLUSION

The relation between the macroscopic- and microscopic-anisotropy parameters is found by the conventional method, using angular derivatives of the free energy. The approximation involved in this procedure is investigated in the spin-wave theory and is found valid when the anisotropy is small compared with the isotropic exchange interaction and when the entropy contribution can be neglected, i.e., at low temperatures. The generally assumed angular dependence of the free energy is only valid within the same limits. A more general spin-wave expression for the free energy is given which is valid for larger anisotropy and higher temperatures.

The precession of the moments in spin waves of systems with large anisotropy is generally nonspherical. The moments tilt less in the anisotropically hard directions than in the easy directions. This has significant effects on the magnitude and temperature dependence of the anisotropy contribution to the free energy (the anisotropy constants) and the resonance energy in particular for the high-order l terms. Large deviations are found from the classical $\frac{1}{2}l(l+1)$ laws and the numerical values are calculated for Gd, Tb, and Dy. It is found that it is valid to parametrize the temperature dependence, using the relative magnitization instead of the temperature.

Qualitatively the effects are physically clear. In the case where the axial anisotropy confines the moments to the basal plane the elliptic precession reduces the temperature dependence of both the macroscopic axial anisotropy and of the axial contribution to the resonance energy. The temperature dependence is, on the other hand, increased both for the basal-plane strain (γ strain) and for the contribution of the basal-plane anisotropy to the resonance energy.

High precision measurements of the magnetization and anisotropy would be valuable for testing the present theory. Existing measurements of the spin waves should be reanalyzed, taking the discussed effects into account.

APPENDIX: RESONANCE ENERGY IN THE CASE OF PLANAR SINGLE-ION ANISOTROPY INCLUDING γ STRAIN

The case where the equilibrium direction of the moment is in the basal plane is important for the heavy rare-earth metals. As discussed in Sec. III all anisotropic terms in (1) can be treated analogously as the single-ion term (3). By rotating (3) into a representation with the quantization axis in the basal plane we obtain the particular form of (25) when the y axis is along the hexagonal axis:

$$H_{\text{an}}=H_{\text{ex}}+\sum_{l,m}K_l^mO_l^m(c)\ ,$$

where

$$\begin{aligned}&K_2^0=-\tfrac{1}{2}B_2^0,\quad K_2^2=-\tfrac{3}{2}B_2^0,\\&K_4^0=\tfrac{3}{8}B_4^0,\quad K_4^2=\tfrac{5}{2}B_4^0,\quad K_4^4=\tfrac{35}{8}B_4^0,\\&K_6^0=-\tfrac{1}{16}(5B_6^0-B_6^6),\quad K_6^2=-\tfrac{15}{32}(7B_6^0+B_6^6),\\&K_6^4=-\tfrac{3}{16}(21B_6^0-B_6^6),\quad K_6^6=-\tfrac{1}{16}(231B_6^0-B_6^6).\end{aligned}\tag{A1}$$

The formula for the resonance energy (33) is then, for $J>1$,

$$\begin{aligned}A_0(T)+B_0(T)=&-(1/J)36S_6B_6^6\\&\times\{1-20[\Delta M(T)+\tfrac{1}{2}b(T)]\}+\delta^+(T)\ ,\\A_0(T)-B_0(T)=&-\frac{2}{J}\sum_l(-1)^{l/2}S_lB_l^0(l-1)\alpha_l\\&\times\{1-(\alpha_l-1)[\Delta M(T)-\tfrac{1}{2}b(T)]\}\\&-(6/J)S_6B_6^6\{1-20[\Delta M(T)-\tfrac{1}{2}b(T)]\}+\delta^-(T)\end{aligned}\tag{A2}$$

$$\begin{aligned}\delta^{\pm}(T)=\pm\frac{1}{2J}\Big(&\sum_l(-1)^{l/2}S_lB_l^0(l-1)\alpha_l\{\Delta M(T)\\&-[(\alpha_l-2)\mp(\alpha_l-3)]b(T)\}\\&-15S_6B_6^6[\Delta M(T)-(6\pm19)b(T)]\Big)\ .\end{aligned}\tag{A3}$$

$\delta^{\pm}$ is small compared with the main terms in (A2). The resonance energy is, according to (31), $\Delta=\{[A_0(T)+B_0(T)][A_0(T)-B_0(T)]\}^{1/2}$. For pure planar anisotropy ($B_6^6=0$) Δ must be zero according to the Goldstone theorem. This theorem is violated in the simple spin-wave theory to first order in $1/J$, ΔM, and b. Brooks Harris[17] has investigated the similar problem of an anisotropic antiferromagnet and shown that Δ vanishes if the dynamical interaction is treated more correctly. We shall therefore neglect $\delta(T)$ in the case of planar anisotropy.

Finally we shall give the explicit expressions for the planar anisotropy case including the γ strain in the frozen-lattice approximation. We find, using (7) and (23), that $\langle\epsilon_1\rangle=\mathcal{C}-\frac{1}{2}\mathcal{A}$ for $\phi=0$, where

$$\mathcal{C}=\frac{1}{c^{\gamma}}[\tfrac{1}{2}B_2^2(\langle O_2^0\rangle-\langle O_2^2\rangle)-\tfrac{1}{8}B_4^2(\langle O_4^0\rangle+4\langle O_4^2\rangle-7\langle O_4^4\rangle)$$

$$-\tfrac{1}{16}B_6^2(\langle O_6^0\rangle+\tfrac{17}{2}\langle O_6^2\rangle+3\langle O_6^4\rangle-\tfrac{33}{2}\langle O_6^6\rangle)] , \tag{A4}$$

$$\mathcal{A}=-\frac{2}{c^{\gamma}}[\tfrac{1}{8}B_4^4(\langle O_4^0\rangle-4\langle O_4^2\rangle+\langle O_4^4\rangle)$$

$$-\tfrac{1}{16}B_6^4(\langle O_6^0\rangle+\tfrac{5}{2}\langle O_6^2\rangle-13\langle O_6^4\rangle+\tfrac{11}{2}\langle O_6^6\rangle)] .$$

We have abbreviated B_{lm}^{γ} by B_l^m as in Ref. 18. If we only consider the terms of lowest order in l we may express the temperature dependence as

$$\mathcal{C}\sim\mathcal{C}_0 m^{\beta_2^s}, \qquad \mathcal{A}\sim\mathcal{A}_0 m^{\beta_4^s} .$$

For Tb and Dy we find from (36) and Table IV $\beta_2^s\sim 3.4$ and $\beta_4^s\sim 14.6$. The result is when the temperature dependence can be described by $m(T)=m$, as for Tb and Dy;

$$A_0(T)+B_0(T)$$
$$=(1/J)[4(c^{\gamma}/J)(\mathcal{C}_0^2 m^{\beta_2^+ +\beta_2^s}+\mathcal{A}_0^2 m^{\beta_4^+ +\beta_4^s})$$
$$-(c^{\gamma}/J)\mathcal{A}_0\mathcal{C}_0(3m^{\beta_4^+ +\beta_2^s}+2m^{\beta_2^+ +\beta_4^s})$$
$$-36B_6^6 S_6 m^{\beta_6^+}] , \tag{A5}$$

$$A_0(T)-B_0(T)$$
$$=(1/J)\{(c^{\gamma}/J)[2\mathcal{C}_0^2 m^{\beta_2^- +\beta_2^s}+\mathcal{A}_0^2 m^{\beta_4^- +\beta_4^s}$$
$$-\mathcal{A}_0\mathcal{C}_0(2m^{\beta_4^- +\beta_2^s}+m^{\beta_2^- +\beta_4^s})]$$
$$+2(3B_2^0 S_2 m^{\beta_2^-}-30B_4^0 S_4 m^{\beta_4^-}$$
$$+105B_6^0 S_6 m^{\beta_6^-})-6B_6^6 S_6 m^{\beta_6^-}+X\} ,$$

where

$$X=-2(7B_4^2 S_4 m^{\beta_4^-}-18B_6^2 S_6 m^{\beta_6^-}$$
$$+11B_6^4 S_6 m^{\beta_6^-})\langle\epsilon_1\rangle=\Delta\mathcal{M}(T)+Y .$$

X shows that the macroscopic magnetostriction parameters do not adequately describe the A-B bracket. An important fact is that X involves a part which transforms under rotation of the average moment in the basal plane as cos6ϕ. This term was first mentioned by Lindgård.[18] It is defined as

$$\Delta\mathcal{M}(T)=(7B_4^2 S_4 m^{\beta_4^-}-18B_6^2 S_6 m^{\beta_6^-})\mathcal{A}_0 m^{\beta_4^s}$$
$$-22B_6^4 S_6\mathcal{C}_0 m^{\beta_6^- +\beta_2^s} . \tag{A6}$$

It is clear from (9) and (20) that also two-ion anisotropy of the appropriate symmetry will contribute to $\Delta\mathcal{M}(T)$. Y is of axial symmetry. The only terms in (A5) transforming under rotation as cos6ϕ are B_6^6, $\mathcal{A}_0\mathcal{C}_0$, and $\Delta\mathcal{M}$; all other terms are invariant. The dominant contribution to $B_0(T)$ in Tb and Dy is the axial anisotropy B_2^0. Therefore the effect of a ϕ dependence of the ellipticity parameter $b(T)$ is not likely to be large, as suggested by Brooks and Egami.[16] Spin-wave measurements on terbium[20] show that $\Delta\mathcal{M}$ is the dominant sixfold term. It is about 20% of the axial anisotropy in magnitude. Since $b(T)$ to a good approximation is proportional to $B_0(T)$, Eq. (32), we expect a 20% variation in $b(T)$ under rotation of the moment direction in the basal plane. The effect is essentially caused by a change in the ground state under the rotation. The results in Table IV can easily be adjusted to take this into account.

ACKNOWLEDGMENTS

We thank A. R. Mackintosh and J. Jensen for critically reading the manuscript.

---

[1] *Magnetic Properties of Rare Earth Metals*, edited by R. J. Elliott (Plenum, London, 1972).

[2] H. B. Callen and E. R. Callen, J. Phys. Chem. Solids 27, 1271 (1966); E. R. Callen and H. B. Callen, Phys. Rev. 129, 578 (1963), *ibid.* 139, A455 (1965).

[3] B. R. Cooper, Phys. Rev. 169, 281 (1968).

[4] P.-A. Lindgård, A. Kowalska, and P. Laut, J. Phys. Chem. Solids 28, 1357 (1967).

[5] M. S. S. Brooks, D. A. Goodings, and H. I. Ralph, J. Phys. C 1, 139 (1968); *ibid.* 2, 1596 (1968); M. S. S. Brooks, Phys. Rev. B 1, 2257 (1970); D. A. Goodings and B. W. Southern, Can. J. Phys. 49, 1137 (1971).

[6] P.-A. Lindgård and O. Danielsen, J. Phys. C 7, 1523 (1974); O. Danielsen, Risø Rept. No. 295 (1974).

[7] B. R. Cooper, R. J. Elliott, S. J. Nettel, and H. Suhl, Phys. Rev. 127, 57 (1952).

[8] R. J. Elliott and K. W. H. Stevens, Proc. R. Soc. A 219, 387 (1953).

[9] O. Danielsen and P.-A. Lindgård, Risø Report No. 259 (1972).

[10] J. Smitt and H. G. Beljers, Philips Res. Rept. 10, 113 (1955).

[11] W. C. Koehler, H. R. Child, R. M. Nicklow, H. G. Smith, R. M. Moon, and J. W. Cable, Phys. Rev. Lett. 24, 16 (1970).

[12] H. B. Møller, J. C. G. Houmann, M. Nielsen, and A. R. Mackintosh, in Ref. 1.

[13] R. M. Niclow and N. Wakabayasi, Proceedings of the 5th IAEA Symposium on Neutron Inelastic Scattering, Grenoble, 1972 (unpublished).

[14] See a review by F. Keffer, in *Handbuch der Physik* (Springer, Berlin, 1966), Vol. XVIII 12, p. 1.

[15] See references in the review by J. J. Rhyne in Ref. 1.

[16] M. S. S. Brooks and T. Egami, J. Phys. C 6, 513 (1972).

[17] A. Brooks Harris, Phys. Rev. Lett. 21, 602 (1968).

[18] P.-A. Lindgård, J. Phys. (Paris) Suppl. 32, 2–3, C1-238 (1971). A number of misprints occur in the intermediate results; they are corrected in this paper.

[19] P.-A. Lindgård, Risø Report No. 313 (1974).

[20] J. G. Houmann, J. Jensen, and P. Touborg (unpublished).

[21] J. Dyson, Phys. Rev. 102, 1217 (1956); 102, 1230 (1956).

[22] P.-A. Lindgård, J. Als-Nielsen, R. J. Birgeneau, and H. J. Guggenheim, Proceedings of the International Conference on Magnetism, Moscow, 1973; J. Phys. C (to be published).