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Theory of the Temperature Dependence of the Spin-Wave Excitation Energies in the Rare-Earth Metals*

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The temperature dependence of the spin-wave excitation energies in the ferromagnetic heavy rare-earth metals, due to spin-wave interactions originating in the strong crystalline electric fields, is studied from both macroscopic and microscopic viewpoints. Consistency between an interacting spin-wave theory, linearized in the Hartree-Fock approximation, and a suitable extension of Smit's macroscopic theory is obtained. Both approaches are consistent with Kanamori and Tachiki's semiphenomenological spin-wave theory. The resonance frequency may be expressed as a geometric mean of the axial and planar anisotropy fields, provided that the low-temperature behavior of the anisotropy constants is modified to allow for elliptical spin precession. At higher temperatures, the results are consistent with Cooper's phenomenological theory. For Tb, agreement with experiment is excellent, but there is a discrepancy in the magnitude of the resonance frequency in Dy.

I. INTRODUCTION

SPIN-WAVE excitations in the heavy rare-earth metals have been the subject of several recent experimental and theoretical studies. Neutron inelastic scattering[1] in Tb and ferromagnetic resonance experiments[2–5] in both Tb and Dy indicate that the spin-wave excitation energies in these metals are strongly temperature-dependent. Early theoretical studies[6] neglected the effects of spin-wave interactions completely. More recently, Cooper[7] has used a phenomenological theory to introduce a temperature dependence into the uniform mode frequency. An earlier attempt[8] to calculate the temperature dependence of the spin-wave spectrum by treating spin-wave interactions explicitly was only partially successful, since several inconsistencies remained in the theory.

The magnetic properties of the heavy rare-earth metals are described by the usual exchange coupling (exchange energy $\sim 10^9$ ergs/cc for Tb) and an extraordinarily strong crystal field. The metals Tb and Dy crystallize in a hcp structure. The largest crystal-field term is the lowest-order axial anisotropy (for Tb $\sim 5\times 10^8$ ergs/cm^3), the sign of which ensures ferromagnetic alignment along a planar easy direction. The other important contributions to the magnetic Hamiltonian are a planar crystal-field term of sixfold symmetry and magnetoelastic effects; it has not been necessary to introduce two-ion anisotropy or higher-order axial crystal-field terms to explain experimental results.

The purpose of the present work is to examine the spin-wave and macroscopic theories to obtain consistent results for the resonance frequency (spin-wave theory is, of course, valid not only for $\mathbf{q}=0$). Kanamori and Tachiki's[9] semiphenomenological spin-wave theory, which is also applied to the problem, appears to be the most convenient way to calculate spin-wave renormalization effects in strong fields.

The organization of this paper is as follows: In Sec. II the macroscopic theory is extended and the resonance frequency so obtained shown to be in agreement with the results of the Kanamori-Tachiki theory. In Sec. III a spin-wave theory based on the Oguchi expansions is examined to illustrate the difficulties associated with this method. An equations-of-motion technique, quite similar to, but for present purposes simpler than, the Green's-function equation of motion method, is then developed and found to be consistent with the Kanamori-Tachiki theory. Kanamori and Tachiki's result is then used to investigate magnetoelastic effects on the spin-wave spectrum (Sec. IV). The results obtained differ from those of Cooper's[7] theory at low temperatures but above temperatures typically ~40–50°K for Dy and Tb there should be no significant difference. One exception, however, is that the expression for the contribution of the frozen lattice term to the resonance frequency differs from Cooper's by a factor of $\frac{2}{3}$. The results of the present work are found to compare favorably with experiment for Tb but there is a sizable discrepancy in the magnitude of the resonance frequency in Dy.

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II. FERROMAGNETIC RESONANCE FREQUENCY

We wish to consider a ferromagnetic system described by the following Hamiltonian:

$$\mathcal{H}=\mathcal{H}_0+[\mathcal{H}_1]_s, \tag{1}$$

where

$$\mathcal{H}_0=-\sum_{i>j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-g\mu_B\sum_i \mathbf{H}\cdot\mathbf{S}_i \tag{2}$$

and

$$\mathcal{H}_1=\sum_i \{V_2{}^0Y_{2,i}{}^0(\omega)+V_6{}^6[Y_{6,i}{}^6(\omega)+Y_{6,i}{}^{-6}(\omega)]\}. \tag{3}$$

The $V_l{}^m$ are crystal-field parameters defined by Elliott and Stevens.[10] $\mathbf{S}_i$ represents the *total* angular momentum on the atom i, therefore the exchange parameter J_{ij} contains a factor $(g-1)^2$ from projecting the spin onto the total angular momentum. The $Y_{l,i}{}^m(\omega)$ are surface spherical harmonics[10] referred to crystal axes and $[\cdots]_s$ denotes the spin equivalent.[11] H is the applied field. The first and second terms in $\mathcal{H}_1$ are the lowest-order axial and planar hexagonal anisotropy, respectively. Several contributions, important in the rare earths, have been excluded from (1) but the physics with which we are concerned is retained—magnetoelastic effects, higher-order axial terms, and anisotropic exchange may be added once the behavior of the simple system is understood.

The spherical harmonics $Y_{l,i}{}^m(\omega)$ may be expressed relative to an axis of quantization which is rotated through Euler angles α, β, and γ from the crystal axes. Since the Euler angles α and β are identical, respectively, with polar angles θ and ϕ, giving the direction of magnetization with respect to crystal axes we may write

$$[\mathcal{H}_1]_s=\sum_i [B_2{}^0\sum_{m'} D_{0m'}{}^{(2)*}(\phi,\theta,0)(O_2{}^{m'})_i$$
$$+B_6{}^6\sum_{m'} (D_{6m'}{}^{(6)*}+D_{-6m'}{}^{(6)*})(O_6{}^{m'})_i], \tag{4}$$

and $B_l{}^m=V_l{}^m\theta_l$, where θ_l is an appropriate reduced matrix element.[11] The $O_l{}^m$ are spin-operator equivalents of the spherical harmonics $Y_l{}^m$ referred to the rotated system[8,12] and the $D_{mm'}{}^{*(l)}$ are rotation matrices.[13]

Zero-temperature spin-wave energies, for an arbitrary axis of magnetization defined by (θ,ϕ), may be derived[14] using the standard Holstein-Primakoff[15] technique; we find[14]

$$\mathcal{H}=C+\sum_{\mathbf{q},i} E_{\mathbf{q}}{}^{(i)}r_{\mathbf{q}}{}^{(i)}, \tag{5}$$

where

$$C=-S^2\sum_{i>j} J_{ij}-g\mu_BH_\phi NS+2NB_2{}^0S(\tfrac{1}{2})P_2{}^0(\cos\theta)$$
$$+NB_6{}^6S(\tfrac{5}{2})\cos6\phi, \tag{6}$$

where

$$S(n)=S(S-\tfrac{1}{2})(S-1)\cdots(S-n) \tag{7}$$

and N is the total number of atoms/cm³. H_ϕ is the component of the applied field along the axis of quantization and

$$E_{\mathbf{q}}{}^{(i)}=\{g\mu_BH_\phi+SJ^{(i)}(\mathbf{q})-B_2{}^0(S-\tfrac{1}{2})$$
$$\times[6P_2{}^0(\cos\theta)+P_2{}^2(\cos\theta)]-36\cos6\phi\, B_6{}^6S(\tfrac{5}{2})/S\}^{1/2}$$
$$\times\{g\mu_BH_\phi+SJ^{(i)}(\mathbf{q})+B_2{}^0(S-\tfrac{1}{2})$$
$$\times[P_2{}^2(\cos\theta)-6P_2{}^0(\cos\theta)]$$
$$-6B_6{}^6\cos6\phi\, S(\tfrac{5}{2})/S\}^{1/2}, \tag{8}$$

where $i=1$, 2 correspond to the optical and accoustic spin-wave modes, respectively (the hcp lattice, which is non-Bravais, may be thought of as two identical interpenetrating hexagonal sublattices).

$$J^{(i)}(\mathbf{q})=J_0-J_{\mathbf{q}}+J_0{}'\pm|J_{\mathbf{q}}{}'|, \tag{9}$$

$$J_q=\sum_{\substack{j\\ \text{same sublattice as } i}} J_{ij}e^{i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)}, \tag{10a}$$

$$J_{\mathbf{q}}{}'=\sum_{\substack{j\\ \text{other sublattice from } i}} J_{ij}e^{i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)}. \tag{10b}$$

The planar term in (8) is evaluated for $\theta=\frac{1}{2}\pi$, since it will enter further calculations only in this configuration.

The phenomenological macroscopic resonance theory developed by Smit and Belgers[16] has been applied by Cooper[7] to obtain the uniform mode frequency

$$E_{\mathbf{q}=0}=[g\mu_B/M(T)](F_{\theta\theta}F_{\phi\phi}-F_{\theta\phi}{}^2)^{1/2}, \tag{11}$$

where $M(T)$ is the magnetization at temperature T and $F_{\theta\theta}$, etc., denote second derivatives of the free energy with respect to angle evaluated at the equilibrium position $\theta=\frac{1}{2}\pi$, $\phi=\phi$ (in resonance experiments an applied field is sometimes used to align the magnetization away from an easy direction in the plane).

Following Cooper[7] we expand the free energy, for zero applied field

$$F=K_1(T)\sin^2\theta+K_3{}'(T)\sin^6\theta\cos6\phi, \tag{12}$$

then, from (11) and (12),

$$E_{\mathbf{q}=0}=[g\mu_B/M(T)][-2K_1(T)]^{1/2}$$
$$\times[-36K_3{}'(T)\cos6\phi]^{1/2}, \tag{13}$$

where $K_3{}'$ has been neglected in comparison with K_1 in the first bracket. The temperature dependence of the uniform mode frequency, due to spin-wave interactions originating in the crystal-field terms, may be obtained

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by using the theory of Callen and Callen[17] for the temperature dependence of the anisotropy constants K_1 and K_3'. This procedure, first used by Cooper,[7] produces results which are in reasonable agreement with experiment. However, the Callen-Callen theory is based on the assumption that the anisotropy may be treated as a first-order perturbation to exchange states which are cylindrically symmetric about the axis of quantization. We wish to consider the excited states of a system described by (1) in which this symmetry is destroyed. At low temperatures, the spin precession becomes elliptical and standard anisotropy theory may not be employed. We examine first a simple extension of the macroscopic theory by reexpressing the free energy

$$F=-(1/\beta)Ln\,\mathrm{Tr}e^{-\beta\mathcal{H}},\quad \beta=1/KT \tag{14}$$

where $\mathcal{H}$ is now given by (5). Then

$$F_{\theta\theta}=\left\langle\frac{\partial^2\mathcal{H}}{\partial\theta^2}\right\rangle+\beta\left[\left\langle\frac{\partial\mathcal{H}}{\partial\theta}\right\rangle^2-\left\langle\left(\frac{\partial\mathcal{H}}{\partial\theta}\right)^2\right\rangle\right], \tag{15}$$

with similar expressions for $F_{\theta\theta}$, $F_{\theta\phi}$.

In the following we neglect the planar anisotropy and applied field in comparison with the axial anisotropy. For the acoustic mode, (15) becomes, for $H=0$,

$$F_{\theta\theta}=-2K_1(T)=6NB_2{}^0S(\tfrac{1}{2})$$
$$\times\left(1-\frac{3}{NS}\sum_{\mathbf{q}}\frac{J^{(1)}(q)+2B_2{}^0(S-\frac{1}{2})}{E_{\mathbf{q}}{}^{(1)}}\langle n_{\mathbf{q}}{}^{(1)}\rangle\right) \tag{16a}$$

and

$$F_{\phi\phi}=-36K_3'(T)\cos6\phi=-36B_6{}^6S(\tfrac{5}{2})\cos6\phi$$
$$\times\left(1-\frac{21}{NS}\sum_{\mathbf{q}}\frac{J^{(1)}(q)+(36/7)B_2{}^0(S-\frac{1}{2})}{E_{\mathbf{q}}{}^{(1)}}\right)\langle n_{\mathbf{q}}{}^{(1)}\rangle, \tag{16b}$$

$$F_{\theta\phi}=0. \tag{16c}$$

The following expression for the magnetization in planar alignment is also useful:

$$\frac{M(T)}{M(0)}\equiv m=\left(1-\frac{1}{NS}\sum_{\mathbf{q}}\frac{3B_2{}^0(S-\frac{1}{2})+J^{(1)}(\mathbf{q})}{E_{\mathbf{q}}{}^{(1)}}\langle n_{\mathbf{q}}{}^{(1)}\rangle\right). \tag{17}$$

The temperature dependence of the anisotropy constants, associated with a Hamiltonian of the form (1), has been derived in a previous publication[14] (to be referred to as I) using Kanamori and Minatono's suggestion[18] that in strong fields the angular deviation from the easy direction, rather that the ratio of anisotropy to exchange, should be used as the expansion parameter in perturbation theory. The temperature dependence of $K_1(T)$, $K_3'(T)$, expressed in the large parentheses of (16), is identical to that obtained for the relevant anisotropy constants in I. Therefore, we may expect that the expression (13) is generally correct, even in strong fields, but that the temperature dependence of the anisotropy constants is given, at low temperatures, by (16) rather than the Callen-Callen theory, according to which[17]

$$K_n(T)=K_n(0)m^{n(2n+1)} \tag{18}$$

at low temperatures, where $m=M(T)/M(0)$ is the reduced magnetization. Comparing (16) and (17) one notices that if $B_2{}^0\gg J^{(1)}(\mathbf{q})$ for all typically excited spin waves, then

$$K_1(T)=K_1(0)m^2,\quad K_3'(T)=K_3'(0)m^{36} \tag{19a}$$

rather than

$$K_1(T)=K_1(0)m^3,\quad K_3'(T)=K_3'(0)m^{21}. \tag{19b}$$

At higher temperatures or for small anisotropy, where $J^{(1)}(\mathbf{q})\gg B_2{}^0$, the temperature dependence expressed in (16) is given by (18) or (19b). A full discussion of equations (19) is given in I. We merely note here that (19a) rather than (19b) is a consequence of the effect of elliptical spin precession on thermal averages; the anisotropy fields in and perpendicular to the basal plane no longer average out equally rapidly.

The frequency of the $\mathbf{q}=0$ mode may also be obtained from the semiphenomenological spin-wave theory developed by Kanamori and Tachiki,[9] who obtain

$$E_{\mathbf{q}=0}=\frac{\{\langle[S_x,[S_x,\mathcal{H}]]\rangle\langle[S_y,[S_y,\mathcal{H}]]\rangle\}^{1/2}}{\langle S_z\rangle}, \tag{20}$$

where $\langle\cdots\rangle$ denotes the thermal average and S_x, S_y, and S_z are components of the total spin when z lies along the magnetization axis. Using (1)–(3), and calculating thermal averages by spin-wave theory [(5)–(7) specialized to $\theta=\frac{1}{2}\pi$], it may easily be shown that (20) is in complete agreement with (13) and (16). Equation (20) will be used again in Sec. IV since this appears to be the simplest way to obtain the resonance frequency for more complex Hamiltonians than (1).

III. SPIN-WAVE RENORMALIZATION

In a previous publication[8] (to be referred to as II) an attempt to calculate the effects of spin-wave interactions was only partially successful. The method used was as follows: The square roots of the Holstein-Primakoff substitutions for spin to Bose operators[15] were expanded and the series truncated so that only quartic terms in Bose operators were retained. The variational principle for the free energy[19] was then used to decouple the quartic terms. As such this was a varia-

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[19] J. M. Radcliffe, Phys. Rev. **165**, 635 (1968).

tional method but the results are completely equivalent to those obtained by symmetric decoupling of the quartic terms.[20] The difficulties associated with this method are indicated by examining the following simple Hamiltonian, similar to (1):

$$\mathcal{H}=-\sum_{i>j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+D\sum_i (S_i^x)^2. \tag{21}$$

Equation (21) is transformed to boson spin-wave operators as described above and the Hamiltonian is diagonalized by the canonical Holstein-Primakoff transformation. Then that part of the Hamiltonian quadratic in quasiparticle operators may be written, for the acoustic mode,

$$\begin{aligned}\mathcal{H}'=\sum_{\mathbf{q}}&[SJ^{(1)}(\mathbf{q})+D(S-\tfrac{1}{2})+DS^{1/2}(S-\tfrac{1}{2})^{1/2}]^{1/2}\\&\times[SJ^{(1)}(\mathbf{q})+D(S-\tfrac{1}{2})-DS^{1/2}(S-\tfrac{1}{2})^{1/2}]^{1/2}n_{\mathbf{q}}.\end{aligned} \tag{22}$$

For $\mathbf{q}=0$ the spin-wave energies are complex indicating an instability in the spin-wave spectrum for the uniform mode. Clearly, $E_{\mathbf{q}=0}$ should simply be zero in the absence of planar anisotropy.[21] Not only is the zero-temperature spin-wave theory unsatisfactory but the finite-temperature results in II, apart from containing similar inconsistencies to those in (22), are not in agreement with the macroscopic theory. It was not possible, therefore, to substitute anisotropy constants into the temperature-dependent spin-wave dispersion relations in the manner described in Sec. II; a result which contradicts physical ideas[22] concerning the effects of anisotropy upon the spin-wave energies.

An expression for the spin-wave energies at finite temperatures, which is more physically reasonable, will now be derived. To simplify the algebra in the first instance, we will examine the simple Hamiltonian (21). Spin-wave operators, attached to the reciprocal lattice, are defined

$$\mathbf{S}_{\mathbf{q}}=(1/N^{1/2})\sum_i \mathbf{S}_i e^{i\mathbf{q}\cdot\mathbf{r}_i} \tag{23}$$

and obey the commutation relations

$$\begin{aligned}[][S_{\mathbf{q}}^z,S_{\mathbf{q}'}^{\pm}]&=\pm(1/N^{1/2})S_{\mathbf{q}+\mathbf{q}'}^{\pm},\\ [S_{\mathbf{q}}^+,S_{\mathbf{q}'}^-]&=(2/N^{1/2})S_{\mathbf{q}+\mathbf{q}'}^z.\end{aligned} \tag{24}$$

$S_i^z|0\rangle=-S|0\rangle$ defines the axis of quantization in the $-z$ direction so that

$$S_i^-|0\rangle=|0\rangle,$$

where $|0\rangle$ is the ground state. N is the number of atoms/cm³ (it will be assumed, for simplicity, that the lattice is Bravais). Equation (21) becomes

$$\begin{aligned}\mathcal{H}=-\sum_{\mathbf{q}}&\tfrac{1}{2}J(\mathbf{q})\{S_{\mathbf{q}}^zS_{-\mathbf{q}}^z+S_{\mathbf{q}}^+S_{-\mathbf{q}}^-\}\\&+(\tfrac{1}{4}D)\sum_{\mathbf{q}}\{S_{\mathbf{q}}^+S_{-\mathbf{q}}^++S_{\mathbf{q}}^-S_{-\mathbf{q}}^-\\&\qquad+S_{\mathbf{q}}^+S_{-\mathbf{q}}^-+S_{\mathbf{q}}^-S_{-\mathbf{q}}^+\}.\end{aligned} \tag{25}$$

We shall also find useful the following expansion for S_i^{z} [23]:

$$\begin{aligned}S_i^z=-S+&\frac{1}{2S}S_i^+S_i^-\\&+\frac{1}{(2S)^2(2S-1)}S_i^+S_i^+S_i^-S_i^-+\cdots\end{aligned} \tag{26}$$

and its Fourier component

$$S_{\mathbf{q}}^z=-S\delta_{\mathbf{q},0}N^{1/2}+\frac{1}{2SN^{1/2}}\sum_{\mathbf{q}'}S_{\mathbf{q}+\mathbf{q}'}^+S_{-\mathbf{q}'}^-+\cdots. \tag{27}$$

The equations of motion for $S_{q_1}^+$, $S_{q_1}^-$ are

$$\begin{aligned}-i\dot{S}_{\mathbf{q}_1}^+&=[\mathcal{H},S_{\mathbf{q}_1}^+]\\&=\frac{1}{N^{1/2}}\sum_{\mathbf{q}}[\mathcal{J}(\mathbf{q},\mathbf{q}_1)S_{\mathbf{q}}^+S_{\mathbf{q}_1-\mathbf{q}}^z\\&\qquad-D(S_{\mathbf{q}_1-\mathbf{q}}^z+\tfrac{1}{2}N^{1/2}\delta_{\mathbf{q},\mathbf{q}_1})S_{\mathbf{q}}^-\\&\qquad\qquad-DS_{\mathbf{q}}^+(S_{\mathbf{q}_1-\mathbf{q}}^z+\tfrac{1}{2}N^{1/2}\delta_{\mathbf{q},\mathbf{q}_1})],\end{aligned} \tag{28}$$

$$\begin{aligned}-i\dot{S}_{\mathbf{q}_1}^-&=[\mathcal{H},S_{\mathbf{q}_1}^-]\\&=-\frac{1}{N^{1/2}}\sum_{\mathbf{q}}[\mathcal{J}(\mathbf{q},\mathbf{q}_1)S_{\mathbf{q}_1-\mathbf{q}}^zS_{\mathbf{q}}^-\\&\qquad-D(S_{\mathbf{q}_1-\mathbf{q}}^z+\tfrac{1}{2}N^{1/2}\delta_{\mathbf{q},\mathbf{q}_1})S_{\mathbf{q}}^-\\&\qquad\qquad-DS_{\mathbf{q}}^+(S_{\mathbf{q}_1-\mathbf{q}}^z+\tfrac{1}{2}N^{1/2}\delta_{\mathbf{q},\mathbf{q}_1})],\end{aligned} \tag{29}$$

where

$$\mathcal{J}(\mathbf{q},\mathbf{q}_1)=J_{\mathbf{q}}-J_{\mathbf{q}_1-\mathbf{q}} \tag{30}$$

and

$$J_{\mathbf{q}}=\sum_j J_{ij}e^{i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)}.$$

Normal ordering of spin operators has been retained in (28) and (29). At zero temperature $S_{\mathbf{q}_1-\mathbf{q}}^z\rightarrow-S\delta_{\mathbf{q}_1,\mathbf{q}}N^{1/2}$. Then

$$\begin{aligned}-i\dot{S}_{\mathbf{q}_1}^+=D(S-\tfrac{1}{2})S_{\mathbf{q}_1}^-&\\+[D(S-\tfrac{1}{2})-S\mathcal{J}(\mathbf{q}_1,\mathbf{q}_1)]S_{\mathbf{q}_1}^+&,\end{aligned} \tag{31}$$

$$\begin{aligned}-i\dot{S}_{\mathbf{q}_1}^-=-D(S-\tfrac{1}{2})S_{\mathbf{q}_1}^+&\\-\{D(S-\tfrac{1}{2})-S\mathcal{J}(\mathbf{q}_1,\mathbf{q}_1)\}S_{\mathbf{q}_1}^-&,\end{aligned} \tag{32}$$

a pair of coupled equations which may easily be solved to yield

$$E_{\mathbf{q}_1}=[-S\mathcal{J}(\mathbf{q}_1,\mathbf{q}_1)]^{1/2}[-S\mathcal{J}(\mathbf{q}_1,\mathbf{q}_1)+2D(S-\tfrac{1}{2})]^{1/2}. \tag{33}$$

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[20] This is just random-phase approximation for the Bose operators; e.g., O. Nagai, Phys. Rev. **180**, 557 (1969).

[21] Complex energies in the zero-temperature theory have not been encountered by previous authors (e.g., Refs. 6 and 7) because their results were carried only to highest order in S.

[22] F. Keffer, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1966), Vol. XVIII/2, p. 73.

[23] M. Wortis, thesis, Harvard University, 1963 (unpublished).

The uniform mode frequency is now zero. Equation (33) also exhibits the property that for spin $\frac{1}{2}$ the effect of axial ansiotropy vanishes, as purely group theoretical considerations would lead one to expect.

We now consider the extension of the theory to finite temperatures, substituting the expansion (27) into the equations of motion for $S_{\mathbf{q}1}{}^{\pm}$. At this stage we separate the effects of spin-wave interactions originating in the exchange term (exchange renormalization) and those originating in the crystal-field terms (crystal-field renormalization) by evaluating the exchange term in (28) and (29) only at zero temperature. Equation (28) becomes

$$\begin{aligned}-i\dot{S}_{\mathbf{q}1}{}^{+}&=D(S-\tfrac{1}{2})S_{\mathbf{q}1}{}^{-}+[D(S-\tfrac{1}{2})-S\mathcal{J}(\mathbf{q}_1,\mathbf{q}_1)]S_{\mathbf{q}1}{}^{+}\\&\quad-\frac{D}{2NS}\sum_{\mathbf{q},\mathbf{q}'}S_{\mathbf{q}}{}^{+}S_{\mathbf{q}1+\mathbf{q}'-\mathbf{q}}{}^{+}S_{-\mathbf{q}'}{}^{-}+\cdots\\&\quad-\frac{D}{2NS}\sum_{\mathbf{q},\mathbf{q}'}S_{\mathbf{q}1+\mathbf{q}'-\mathbf{q}}{}^{+}S_{-\mathbf{q}'}{}^{-}S_{\mathbf{q}}{}^{-}+\cdots,\end{aligned}\tag{34}$$

with a similar expression for $S_{\mathbf{q}1}{}^{-}$. The equations of motion are now linearized in the following manner: Fluctuation parts of the three operator terms plus all the higher-order terms (i.e., terms in five, seven operators, etc.) are neglected. Then the triple operator terms are symmetrically decoupled, resulting in a temperature-dependent Hartree-Fock approximation. Noticing from (32) that a linear combination of the operators $S_{\mathbf{q}}{}^{+}$ and $S_{\mathbf{q}}{}^{-}$ generates excited states in the sense that if $T_{\mathbf{q}}{}^{+}$ is such a combination, then $[\mathcal{H},T_{\mathbf{q}}{}^{+}]=E_{\mathbf{q}}T_{\mathbf{q}}{}^{+}$, we recognize that the expectation values of $S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{+}$, $S_{\mathbf{q}}{}^{-}S_{-\mathbf{q}}{}^{-}$ and $S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}$ are nonzero. Defining $\langle\cdots\rangle$ as the thermal average over an effective single-particle density matrix, to be determined self-consistently, the linearized coupled equations may be written

$$-i\dot{S}_{\mathbf{q}}{}^{+}=A_{\mathbf{q}}(T)S_{\mathbf{q}}{}^{+}+B_{\mathbf{q}}(T)S_{\mathbf{q}}{}^{-},\tag{35a}$$

$$-i\dot{S}_{\mathbf{q}}{}^{-}=-A_{\mathbf{q}}(T)S_{\mathbf{q}}{}^{-}-B_{\mathbf{q}}(T)S_{\mathbf{q}}{}^{+},\tag{35b}$$

where

$$A_{\mathbf{q}}=-S\mathcal{J}(\mathbf{q},\mathbf{q})+B(T),\tag{36a}$$

$$B_{\mathbf{q}}=B(T),\tag{36b}$$

and

$$B(T)=D(S-\tfrac{1}{2})-\frac{D}{2NS}\sum_{\mathbf{q}'}\langle S_{\mathbf{q}'}{}^{+}S_{-\mathbf{q}'}{}^{+}\rangle-\frac{D}{NS}\sum_{\mathbf{q}'}\langle S_{\mathbf{q}'}{}^{+}S_{-\mathbf{q}'}{}^{-}\rangle.\tag{37}$$

The linear combinations

$$\begin{aligned}T_{\mathbf{q}}{}^{+}&=\lambda_{\mathbf{q}}S_{\mathbf{q}}{}^{+}+\mu_{\mathbf{q}}S_{\mathbf{q}}{}^{-},\\T_{\mathbf{q}}{}^{-}&=\lambda_{\mathbf{q}}{}^{+}S_{\mathbf{q}}{}^{-}+\mu_{\mathbf{q}}{}^{+}S_{\mathbf{q}}{}^{+}\end{aligned}\tag{38}$$

diagonalize Eqs. (35) in the sense that $[\mathcal{H},T_{\mathbf{q}}{}^{+}]=E_{\mathbf{q}}\,T_{\mathbf{q}}{}^{+}$. Then the spin-wave energies are

$$\begin{aligned}E_{\mathbf{q}}&=(A_{\mathbf{q}}{}^{2}-B_{\mathbf{q}}{}^{2})^{1/2}\\&=[-S\mathcal{J}(\mathbf{q},\mathbf{q})]^{1/2}[-S\mathcal{J}(\mathbf{q},\mathbf{q})+2B(T)]^{1/2}.\end{aligned}\tag{39}$$

Using (35) and (38) it may easily be shown that

$$\begin{aligned}\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{+}\rangle=\langle S_{\mathbf{q}}{}^{-}S_{-\mathbf{q}}{}^{-}\rangle&=\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle\frac{2\lambda_{\mathbf{q}}\mu_{\mathbf{q}}}{\lambda_{\mathbf{q}}{}^{2}+\mu_{\mathbf{q}}{}^{2}}\\&=-\frac{B_{\mathbf{q}}(T)}{A_{\mathbf{q}}(T)}\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle.\end{aligned}\tag{40}$$

Substituting (40) into (37), we find

$$B(T)=D(S-\tfrac{1}{2})-\frac{D}{2NS}\sum_{\mathbf{q}}\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle\left(2-\frac{B_{\mathbf{q}}(T)}{A_{\mathbf{q}}(T)}\right)\tag{41}$$

and from (27)

$$\frac{M(T)}{M(0)}\equiv m=1-\frac{1}{2NS^{2}}\sum_{\mathbf{q}}\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle+\cdots.\tag{42}$$

By comparing (39), (41), and (42) with (16) and (17) it may be shown that (39) reduces to

$$E_{\mathbf{q}}=[-S\mathcal{J}(\mathbf{q})]^{1/2}\left(-S\mathcal{J}(\mathbf{q})-\frac{2K_1(T)}{NSm}\right)^{1/2}.\tag{43}$$

The extension of the process to include the planar anisotropy is straightforward. If (4) is particularized to $\theta=\frac{1}{2}\pi$, the system is described by the Hamiltonian

$$\begin{aligned}\mathcal{H}=&-\sum_{i>j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-g\mu_B\sum_i\mathbf{H}\cdot\mathbf{S}_i+D\sum_i(S_i{}^{x})^2\\&+B_6{}^{6}\cos6\phi\sum_i[(S_i{}^{z})^6-15(S_i{}^{z})^4(S_i{}^{y})^2\\&+15(S_i{}^{z})^2(S_i{}^{y})^4+\cdots],\end{aligned}\tag{44}$$

where

$$D=\tfrac{3}{2}B_2{}^{0}.\tag{45}$$

If (21) is replaced by (44), the procedure outlined from (23)–(35) leads to a pair of coupled equations of the same form as (35) with $A_{\mathbf{q}}$, $B_{\mathbf{q}}$ redefined as

$$\begin{aligned}A_{\mathbf{q}}&=-S\mathcal{J}(\mathbf{q},\mathbf{q})+B(T)+C(T),\\B_{\mathbf{q}}&=B(T)+D(T),\end{aligned}\tag{46}$$

with $B(T)$ given by (37), and

$$\begin{aligned}C(T)=B_6{}^{6}\cos6\phi\,S(\tfrac{5}{2})/S\{21+2NS^{-2}[&-420\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle\\&+150\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{+}\rangle+135\langle S_{\mathbf{q}}{}^{-}S_{-\mathbf{q}}{}^{-}\rangle]\},\end{aligned}\tag{47}$$

$$\begin{aligned}D(T)=B_6{}^{6}\cos6\phi S(\tfrac{5}{2})/S\{-15+2NS^{-2}[&-300\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{-}\rangle\\&-45\langle S_{\mathbf{q}}{}^{+}S_{-\mathbf{q}}{}^{+}\rangle-210\langle S_{\mathbf{q}}{}^{-}S_{-\mathbf{q}}{}^{-}\rangle]\}.\end{aligned}\tag{48}$$

$S(\frac{5}{2})$ is defined by (7). Following the procedure outlined in (38)–(43) we obtain the spin-wave excitation energies

$$E_{\mathrm{q}}=\Bigg[-S\mathcal{J}(\mathbf{q},\mathbf{q})-36\cos 6\phi\, B_6{}^6S(\tfrac{5}{2})/S$$

$$\times\left(1-\frac{20\langle S_{\mathrm{q}}{}^+S_{-\mathrm{q}}{}^-\rangle-15\langle S_{\mathrm{q}}{}^+S_{-\mathrm{q}}{}^+\rangle}{NS^2}\right)\Bigg]^{1/2}$$

$$\times\Bigg[-S\mathcal{J}(\mathbf{q},\mathbf{q})+2D(S-\tfrac{1}{2})$$

$$\times\left(1-\frac{2\langle S_{\mathrm{q}}{}^+S_{-\mathrm{q}}{}^-\rangle+\langle S_{\mathrm{q}}{}^+S_{-\mathrm{q}}{}^+\rangle}{2NS^2}\right)\Bigg]^{1/2}$$

$$=\left(-S\mathcal{J}(\mathbf{q},\mathbf{q})-\frac{36K_3{}'(T)\cos 6\phi}{NSm}\right)^{1/2}$$

$$\times\left(-S\mathcal{J}(\mathbf{q},\mathbf{q})-\frac{2K_1(T)}{NSm}\right)^{1/2},\qquad(49)$$

where $K_1(T)$ and $K_3{}'(T)$ are given by (16) and $\mathcal{J}(\mathbf{q},\mathbf{q})$ is defined by (30).

IV. MAGNETOELASTIC EFFECTS ON SPIN-WAVE SPECTRUM

The Hamiltonian (1) is not sufficient to explain recent neutron scattering and resonance experiments.[1–5] The experimentally observed energy gap for the acoustic mode is several times larger than the value estimated when the experimentally observed quantities K_1 and $K_3{}'$ are inserted into (13). This anomaly has been studied by Cooper,[7] who introduced magnetoelastic effects into noninteracting spin-wave theory. Cooper found that the discrepancy in the size of the energy gap might be removed by including a magnetoelastic term which dominates the planar anisotropy in what is known as the "frozen lattice approximation."[24] However, part of the temperature dependence of this term is neglected in noninteracting spin-wave theory; an approximation which is redeemed only by inconsistent definitions of the magnetoelastic coupling constants. Here we reexamine the spin-wave excitation energies in the frozen lattice approximation. Specifically, we follow Cooper's use of the Callen-Callen presentation of magnetostriction theory, but use the Kanamori-Tachiki result (20) to obtain the temperature-dependent resonance frequency.

For a ferromagnet in planar alignment the lowest-order single-ion magnetoelastic contribution to H is given, in the Callen-Callen notation[25] by

$$\mathcal{H}_{\mathrm{me}}=-\sum_i(\tfrac{1}{2})\tilde{B}^\gamma\{\epsilon_1{}^\gamma[(S_i{}^x)^2-(S_i{}^y)^2]$$
$$+\epsilon_2{}^\gamma[S_i{}^xS_i{}^y+S_i{}^yS_i{}^x]\}.\qquad(50)$$

The $\epsilon_i{}^{(\mu,j)}$ are irreducible strains for hcp symmetry and $\tilde{B}^\gamma$ is a magnetoelastic coupling constant which is, by definition, temperature-independent. The elastic contribution is

$$\mathcal{H}_e=\tfrac{1}{2}C^\gamma[(\epsilon_1{}^\gamma)^2+(\epsilon_2{}^\gamma)^2];\qquad(51)$$

C^γ is a phenomenological elastic constant. The z axis in (50) is taken along the c axis and x and y are along and perpendicular to one of the six axes directed toward a neighboring atom in the basal plane, respectively. The equilibrium strains are found by minimizing the free energy with respect to strain:

$$\epsilon_1{}^\gamma=\sum_i\tfrac{1}{2}[(S_i{}^x)^2-(S_i{}^y)^2]\tilde{B}^\gamma/C^\gamma,\qquad(52a)$$

$$\epsilon_2{}^\gamma=\sum_i\tfrac{1}{2}(S_i{}^xS_i{}^y+S_i{}^yS_i{}^x)\tilde{B}^\gamma/C^\gamma.\qquad(52b)$$

If the equilibrium strains follow the instantaneous direction of spin throughout resonance (52) may be substituted into (50), then

$$\mathcal{H}_{\mathrm{me}}=(1/4C^\gamma)(\tilde{B}^\gamma)^2[(S_i{}^x)^2+(S_i{}^y)^2]^2,\qquad(53)$$

$\mathcal{H}_{\mathrm{me}}$ is cylindrically symmetric about the c axis and has no significant effect upon the spin-wave energies.

In the frozen lattice approximation the strains are frozen at their equilibrium positions. The $\epsilon_1{}^\gamma$ and $\epsilon_2{}^\gamma$ are determined by taking the thermal average of the spin-operator expressions in (52). We let the direction of magnetization make the angle ϕ with one of the six axes directed towards a neighboring atom in the basal plane and perform a coordinate transformation so that the axis of magnetization becomes the z axis, then

$$\epsilon_1{}^\gamma=\sum_i\tfrac{1}{2}(\tilde{B}^\gamma/C^\gamma)\langle(S_i{}^z)^2-(S_i{}^y)^2\rangle\cos 2\phi,\qquad(54a)$$

$$\epsilon_2{}^\gamma=\sum_i\tfrac{1}{2}(\tilde{B}^\gamma/C^\gamma)\langle(S_i{}^z)^2-(S_i{}^y)^2\rangle\sin 2\phi.\qquad(54b)$$

The relationship between the experimentally determined magnetostriction coefficient and the equilibrium strains is given by

$$\epsilon_1{}^\gamma=\tfrac{1}{2}\lambda^\gamma\cos 2\phi,\qquad(55a)$$

$$\epsilon_2{}^\gamma=\tfrac{1}{2}\lambda^\gamma\sin 2\phi,\qquad(55b)$$

which for our purposes may be regarded as a definition of the magnetostriction coefficient. Thus using (54) and (55), (50) becomes

$$\mathcal{H}_{\mathrm{me}}=\tfrac{1}{4}\tilde{B}^\gamma\lambda^\gamma\sum_i[(S_i{}^y)^2-(S_i{}^z)^2],\qquad(56)$$

which corresponds to the expression obtained by Cooper. At this stage Cooper uses noninteracting spin-wave theory to calculate the resonance frequency. The λ^γ produces part of the temperature dependence of (56), but the spin-operator expression makes no temperature-dependent contribution in the simple spin-wave approximation. Cooper, however, uses the following ex-

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[24] E. A. Turov and V. G. Shavrov, Fiz. Tverd. Tela **7**, 217 (1965) [English transl.: Soviet Phys.—Solid State **7**, 166 (1965)].

[25] E. Callen and H. B. Callen, Phys. Rev. **139**, A455 (1965).

pression for $\tilde{B}^{\gamma}$:

$$\tilde{B}^{\gamma}=3C^{\gamma}\lambda^{\gamma}/S(2S-1) \tag{57}$$

at finite temperatures [although $\tilde{B}^{\gamma}$, by (50), is temperature-independent], and in this way retrieves the temperature-dependent contributions to the resonance frequency of the spin-operator expression in (56). Comparison of (54) and (55) shows that even at zero temperature (57) is incorrect by a factor of approximately $\frac{3}{2}$, an error which appears to derive from inconsistent definitions of either the magnetostriction coefficients or magnetoelastic coupling constants.

The contribution of (56) to the resonance frequency may be obtained from (20), by noting that

$$\langle[S_x,[S_x,\mathcal{H}_{\text{me}}]]\rangle=\tilde{B}^{\gamma}\lambda^{\gamma}\langle\sum_i\{(S_i^{y})^2-(S_i^{z})^2\}\rangle; \tag{58}$$

hence, using (54), (55), and (49)

$$E_{\mathbf{q}=0}=\frac{[-36K_3'(T)\cos6\phi+C^{\gamma}(\lambda^{\gamma})^2]^{1/2}[-2K_1(T)]^{1/2}}{NSm}. \tag{59}$$

The correlation function $\langle(S_i^{z})^2-(S_i^{y})^2\rangle$ is similar to the correlation function $\langle(S_i^{z})^2-\frac{1}{3}S(S+1)\rangle$ defined by Callen and Callen.[25] When $\langle S_x^2\rangle=\langle S_y^2\rangle$, which is the case for weak anisotropy, the two correlation functions differ only by a constant. In this case, since[25]

$$\langle(S_i^{z})^2-\tfrac{1}{3}S(S+1)\rangle \sim\hat{I}_{5/2}[\mathfrak{L}^{-1}(m)]\sim m^3 \quad\text{as}\quad T\rightarrow0, \tag{60}$$

we have

$$[\lambda^{\gamma}(T)/\lambda^{\gamma}(0)]^2\sim m^6. \tag{61}$$

At low temperatures, subject to the same conditions that the anisotropy constants K_1 and K_3' deviate from the conventional power laws, we find

$$[\lambda^{\gamma}(T)/\lambda^{\gamma}(0)]^2\sim m^8, \tag{62}$$

where the correlation function in (54) was evaluated in the spin-wave approximation.

The spin-wave theory in Sec. III may be extended to include the two spin-wave modes in the non-Bravais lattice (the necessary transformations appear in II). For completeness, therefore, the spin-wave energies may be written

$$\begin{aligned}E_{\mathbf{q}}^{(i)}=&[g\mu_BH_{\phi}+SJ^{(i)}(\mathbf{q})\\&-36K_3'(T)\cos6\phi/NSm+C^{\gamma}(\lambda^{\gamma})^2/NSm]^{1/2}\\&\times[g\mu_BH_{\phi}+SJ^{(i)}(\mathbf{q})-2K_1(T)/NSm]^{1/2},\end{aligned} \tag{63}$$

where $i=1$, 2 correspond to the acoustic and optical spin-wave modes and $J^{(i)}(\mathbf{q})$ is given by (9). H_{ϕ} is the component of the applied field along the equilibrium direction. Applied field effects are discussed in detail by Cooper.[7]

V. COMPARISON WITH EXPERIMENT

In Secs. II and III it was assumed that the planar anisotropy arose solely from the crystal field in the unstrained crystal. Cooper[7] has estimated that the contribution due to second-order hexagonally symmetric magnetostriction effects should also be important in Tb and Dy. The temperature dependence of the magnetostrictive contribution is quite different from that of the unstrained crystal-field term but it has not been possible to determine the origin of the planar anisotropy from experiment unambiguously.[7,14] Since the torque experiments measure the total contribution to the planar anisotropy, we may avoid this problem by using the experimental values for the anisotropy constants to predict the resonance frequency. There are several sets of experimental data available for the anisotropy constants.[26–29] The zero-temperature values are shown in Table I. Also shown are the estimated values of $C^{\gamma}[\lambda^{\gamma}(0)]^2$, the frozen lattice contribution (these values are approximately $\frac{2}{3}$ of those quoted by Cooper).

In the absence of the frozen lattice term the resonance frequency may be reduced to zero by application of a suitable resonance field along a planar hard direction.[7] However, the frozen lattice contribution is isotropic. Therefore, as Turov and Shavrov[24] and Cooper[7] have noted, the most striking effect of the frozen lattice approximation is the appearance of a minimum resonance frequency. Also, in the presence of a dominant frozen lattice term the resonance field increases with increasing temperature, whereas if the resonance field is used to reduce the effect of planar anisotropy it may be expected to decrease with increasing temperature. Furthermore, the frozen lattice term falls off far less rapidly with increasing temperature than the contribution from planar anisotropy, a characteristic which is essential to the explanation of neutron scattering and for infrared resonance results in terbium.

TABLE I. Spin-wave energy parameters from dc measurements at 0°K in 10^8 ergs/cm^3.

	$\frac{2}{3}K_1(0)$	$K_3'(0)$	$C^{\gamma}[\lambda^{\gamma}(0)]^2$
Terbium	5.65[a]	0.0185[a]	0.115[b]
	5.50[c]	0.0242[c]	
	4.50[d]	...	
Dysprosium	5.50[a]	0.11[a]	0.093[b]
	5.50[c]	0.075[c]	
	4.90[d]	...	
	...	0.105[e]	

[a] Reference 29.
[b] Calculated, using same parameters as Cooper (Ref. 7).
[c] Reference 26.
[d] Reference 27.
[e] Reference 48.

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[26] J. J. Rhyne and A. E. Clark, J. Appl. Phys. **38**, 1379 (1967).

[27] J. J. Rhyne, S. Foner, E. J. McNiff, Jr., and R. Doclo, J. Appl. Phys. **39**, 892 (1968).

[28] S. H. Liu, D. R. Behrendt, S. Legvold, and R. H. Good, Jr., Phys. Rev. **116**, 1464 (1959).

[29] J. L. Feron and R. Panthenet, in Rare Earth Conference, Grenoble, 1968 (unpublished).

From Table I it may be seen that in the absence of an applied field the spin-wave gap in Tb and Dy is over 400 GHz at zero temperature. By applying a field along a hard direction Rossel and Jones,[3] and Bagguley and Leisegang[2] have reduced the resonance frequency to 37.7 and 9.44 GHz, respectively. In both cases the signal frequency lies well below the minimum frequency allowed in the frozen lattice approximation, and the resonance field decreases with increasing temperature. An analysis of the Bagguley and Leisegang data in II, and of both sets of data by Cooper, show that the temperature variation of the resonance field may be explained by the presence of only the hexagonal anisotropy in the first bracket of (63), but rather larger values of $K_3'(0)$ than those given in Table I are required for Tb.

The resonance experiments of Wagner and Stanford,[5] and Marsh and Sievers,[4] together with the neutron scattering experiments of Møller, Houmann, and MacKintosh[1] fall into a different category. Wagner and Stanford find that to maintain a resonance frequency of 98.2 GHz in the temperature range 160–240°K, the resonance field in terbium increases with increasing temperature. This result is explained by the presence of the frozen lattice term. Wagner and Stanford find that their data are in reasonable agreement with (63). Using K_1 and $C^\gamma(\lambda^\gamma)^2$ as parameters, they obtain a good fit to their data with $K_1(0)/NS=-18$°K/atom, $C^\gamma(\lambda^\gamma)^2/NS=1.7$°K/atom, compared with 32°K/atom and 2.6°K/atom, respectively, from Table I. The difference may be attributed to the rather large spread in the resonance data.

Marsh and Sievers have measured the resonance frequency, at low temperatures, in zero applied field for both Tb and Dy. Their results for terbium are shown in Fig. 1. Also shown is the theoretical curve from (63) with values of K_1, K_3' taken from Rhyne and Clark's measurements; $C^\gamma[\lambda^\gamma(0)]^2$ is from Table I with temperature dependence given by (61). The anisotropy and resonance experiments are therefore consistent [Marsh and Sievers, who analyze their own data treating K_1, K_3' and $C^\gamma(\lambda^\gamma)^2$ as parameters, obtain slightly different values from those given here]. The situation for dysprosium is less satisfactory. Marsh and Sievers, again using K_1, K_3', and $C^\gamma(\lambda^\gamma)^2$ as parameters, are able to fit the data only if $K_3'\simeq 1.8\times10^7$ ergs/cm^3, i.e., about 70% larger than the largest value in Table I. From Feron's measurements for K_1 and K_3' which give the largest value of the resonance frequency from static measurements, one finds $E_{\mathbf{q}=0}=28.2$°K at zero temperature, compared with Marsh and Sievers's measurement of 35°K.

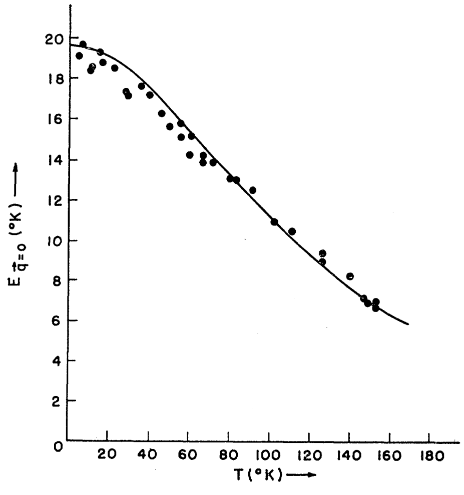

FIG. 1. Temperature dependence of the spin-wave gap in terbium: Experiment: Marsh and Sievers; solid line: from Eq. (63) with K_1 and K_3' taken from the experiments of Rhyne and Clark.

The magnitude of the spin-wave gap in Tb from neutron scattering experiments[1] is quite consistent with the frozen lattice approximation. The spin-wave dispersion relations are not measured at zero $\mathbf{q}$ and extrapolation is difficult at high temperatures when the gap is small. A reliable estimate may be made at 90°K, however, when $E_{\mathbf{q}=0}\simeq 12.9$°K (corresponding to 269 GHz) compared with the theoretical prediction of 12.3°K.

The most noticeable feature of the foregoing is that if the resonance frequency is high ($\gtrsim 100$ GHz) the frozen lattice approximation appears to apply. But if the resonance frequency is lowered sufficiently ($\lesssim 40$ GHz) by application of a suitable resonance field, the frozen lattice contribution disappears. Apparently the lattice may adjust to the instantaneous position of spin throughout resonance below 40 and not above 100 GHz. Even if this is accepted there remains the discrepancy in the magnitude of the anisotropy required to explain both the resonance frequency of Bagguley and Leisegang's experiment for Tb and Marsh and Seivers's result for Dy.

VI. DISCUSSION

The failure of the boson spin-wave theory, outlined in Sec. III, is not confined to a method based on the Oguchi expansion. Under the Dyson-Mallev transformation the Hamiltonian (21) becomes non-Hermitian but may be diagonalized by a nonunitary canonical transformation. This procedure produces the same results as the Oguchi method, as might be expected, since $\mathcal{H}_{\text{Oguchi}}=\frac{1}{2}(\mathcal{H}+\mathcal{H}^\dagger)_{\text{D-M}}$.

It is worth noting that if S_z in Eqs. (28) and (29) is decoupled directly—the usual random-phase approximation—there results a high-temperature approximation. Then Eq. (63) is still valid, since the temperature dependence of the anisotropy constants is given, not by (19), but by the high-temperature limit of the Callen-Callen theory $K_n(T)=K_n(0)m^n$.

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