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Magnetoelastic Spin Hamiltonians: Applications to Garnets

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The theory of magnetoelastic spin Hamiltonians and their relation to magnetostriction is reviewed, clarified, and expanded. We consider terms representing strain modulation of bilinear exchange, of crystal field, and of the **g** tensor (direct-forced magnetostriction). When the unstrained spin Hamiltonian contains anisotropic exchange and large crystal-field terms, we expect some new effects: Strain modulation of the Dzyaloshinski-Moriya interaction can be important for most site symmetries; second-degree terms in spin operators give rise to high-order magnetostriction constants, and high-degree spin terms contribute to low-order magnetostriction; the temperature and field dependences may deviate from the isotropic theory. Application is made to the much-studied Yb^{3+} ion in ytterbium iron garnet and also to a model for dysprosium iron garnet. For the latter case, we show that the novel behavior of the observed magnetostriction can be explained by the large crystal fields.

I. INTRODUCTION

THE original theory of magnetostriction[1] was purely a symmetry argument.[2] The gross distortion of a crystal which depends upon the direction and magnitude of its magnetization was related to a magnetoelastic free energy. This in turn was expanded in such combinations of direction cosines of magnetization and of strain components as were allowed by the crystal-point group. The number of allowed coefficients, that is, the number of magnetostriction constants, could thus be accounted for, but such a theory did not address itself to the temperature dependence of the strain or the origin of the constants.

In the next advance,[3] the phenomenology was directed to the Hamiltonian. Strains were now coupled to appropriate combinations of spin operators, again with assumed expansion constants. Statistical mechanics then yielded the temperature and magnetic field-dependence of those microscopic magnetostriction constants which had been entirely arbitrary in the original work.

Concurrent with the statistical theories, and often intertwined, were the several efforts to derive magnetoelastic coupling constants from their original quantum-mechanical sources. These efforts[4] consisted of an evaluation of the strain dependence of the quantum-mechanical energy, usually based on a point-charge model.

The most recent advances in our understanding of magnetostriction—advances which are the spur for the present paper—are of an experimental nature. The new techniques of electron-paramagnetic resonance (EPR) under pressure,[5] ferrimagnetic,[6] and optical studies[7] under pressure, and of acoustic-paramagnetic resonance[8] (APR) allow direct experimental determination of some of the strain-dependent terms in the spin Hamiltonian. It is the purpose of this paper to clarify the relationship of these terms to magnetostriction.

In Sec. II we discuss some general features of the magnetoelastic spin Hamiltonian. For the simplest forms of this Hamiltonian, we display the symmetry properties for each of the 32 point groups. We show how strain derivatives of a Dzyaloshinski-type interaction can occur. Next we particularize the theory to the case of a Kramers doublet, Yb^{3+} in the iron garnet. This case has already been treated by Phillips and White,[5] but there are a number of ways in which our treatment differs from theirs. In Sec. IV we explore the temperature dependence of the magnetization and magnetostriction of ytterbium iron garnet (YbIG) and of dysprosium iron garnet (DyIG). Previous treatments of the statistical mechanics of magnetostriction minimized crystal-field effects. When anisotropic exchange or crystal fields are large, we shall see that possibilities proliferate for the temperature dependence of the various magnetostriction coefficients.

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II. MAGNETOELASTIC SPIN HAMILTONIAN

A. General Theory

A spin Hamiltonian is a useful shorthand for the interactions within a subspace of the full Hamiltonian. It describes the separations in energy of a set of levels in terms of polynomials in a fictitious spin S, whose multiplicity is equal to the number of levels to be fitted. Levels of arbitrary separation can be fitted by polynomials up to degree $2S$, but often only terms of low degree suffice. When the significant level splittings come from the external field, the exchange interaction, and the crystal field, it is often adequate to employ the spin Hamiltonian

$$\mathcal{H}=\mathbf{H}\cdot\mathbf{g}\cdot\mathbf{S}+\mathbf{H}_{\mathrm{ex}}\cdot\mathbf{g}\cdot\mathbf{S}+\mathbf{S}\cdot\mathbf{D}\cdot\mathbf{S}. \tag{2.1}$$

Here H is the external field (given in energy units), g is the paramagnetic spectroscopic splitting factor, and $\mathbf{H}_{\mathrm{ex}}$ is the effective exchange field on the ion of fictitious spin $\mathbf{S}$, in the molecular field approximation. The third term in Eq. (2.1) represents the effect of the crystal field. For a general fictitious spin, there can be higher-degree terms in Eq. (2.1) in the exchange interaction, and especially in the crystal-field contribution, but we limit ourselves to the simple case, noting that both g and D are often anisotropic tensors. Furthermore, in the molecular-field approximation, the exchange field is itself related to the magnetization by an (often anisotropic) tensor,

$$\mathbf{H}_{\mathrm{ex}}=\mathbf{M}\cdot\boldsymbol{\lambda}. \tag{2.2}$$

We combine λ and g and rewrite the second term of Eq. (2.1) as

$$\mathcal{H}^{\mathrm{ex}}=\mathbf{M}\cdot\mathbf{g}^{\mathrm{ex}}\cdot\mathbf{S}. \tag{2.3}$$

When the crystal strains, g, g^{ex}, and D all change. Expanding to first order in a Taylor series, the magnetoelastic perturbation becomes

$$\mathcal{H}^{\mathrm{me}}=\mathbf{H}\cdot\mathbf{F}\cdot\mathbf{S}:\boldsymbol{\varepsilon}+\mathbf{M}\cdot\mathbf{F}^{\mathrm{ex}}\cdot\mathbf{S}:\boldsymbol{\varepsilon}+\mathbf{S}\cdot\mathbf{G}\cdot\mathbf{S}:\boldsymbol{\varepsilon}, \tag{2.4}$$

where $\mathbf{F}$, $\mathbf{F}^{\mathrm{ex}}$, and $\mathbf{G}$ are fourth-rank tensors, with elements

$$F_{ijkl}=\frac{\partial g_{ij}}{\partial\epsilon_{kl}},\quad F_{ijkl}{}^{\mathrm{ex}}=\frac{\partial g_{ij}{}^{\mathrm{ex}}}{\partial\epsilon_{kl}},\quad G_{ijkl}=\frac{\partial D_{ij}}{\partial\epsilon_{kl}}, \tag{2.5}$$

and ϵ_{kl} are the components of the strain tensor as defined by Sommerfeld. Later we will introduce the Voigt notation and employ the e_i strains of Love as shown by Kittel.[9a]

The three terms in (2.4) may be interpreted as follows. The $\mathbf{F}$ term is a strain-dependent g shift resulting in a "direct" force magnetostriction.[9b] Unlike the usual forced magnetostriction, which comes about because of the susceptibility of the spin-expectation value, this effect should be largest at low temperature. Next, $\mathbf{F}^{\mathrm{ex}}$ gives rise to magnetostriction as the crystal trades elastic energy in order to enhance its energy-reducing exchange interaction. The $\mathbf{G}$ term comes about because the effect of any strain-induced potential depends on the orientation of the charge cloud and is thus a function of $\mathbf{S}$.

As to which terms are important, it is clear that $\mathbf{G}$ is absent for Kramers doublets, $\mathbf{F}$ requires a large external field, and $\mathbf{F}^{\mathrm{ex}}$ needs exchange interactions.

Beyond this, the determination of contributions depends on experiment since nothing more of a general nature is known. This statement applies to S-state ions, about which there seems to be some confusion. For S-state ions all anisotropic effects are small since the ground state is orbitally symmetric in lowest order; any effect of strain must come via excited terms and configurations. The problem of accounting for contributions is not yet resolved for either transition metals (Fe^{3+}, Mn^{2+})[4,10] or rare earths (Gd^{3+}, Eu^{2+}).[11] Thus, for now, there is *no* theoretical reason to believe that crystal-field-type terms ($\mathbf{G}$) are favored over direct ($\mathbf{F}$) and exchange ($\mathbf{F}^{\mathrm{ex}}$) terms in this case. Experimental evidence supports this view: For GdIG[2] the magnetostriction temperature dependence is typical of a dominant $\mathbf{F}^{\mathrm{ex}}$ term. As for the transition metals,[5,12a] the g shifts, which could be interpreted as giving rise to $\mathbf{F}$ and part of $\mathbf{F}^{\mathrm{ex}}$, are known to be unmeasurably small in external fields of the order of 10 kOe. for Mn^{2+} and Fe^{3+} in diamagnetic MgO.[5] However, when we multiply the upper limit on Δg by typical exchange fields, it turns out that this part of the $\mathbf{F}^{\mathrm{ex}}$ term could be much larger than the $\mathbf{G}$ contribution in a ferromagnet. Thus in S-state ions, all terms are small, but none are manifestly smaller than others.

Another note concerns terms higher order in the spin not included in Eq. (2.4). For an isotropic unstrained Hamiltonian, these will contribute only to high-order magnetostriction and only be considerable at low temperature. But in the general case, high-order terms do contribute even to second-order magnetostriction. Mindful of this fact, we exclude them to simplify the discussion.

The number of independent elements in $\mathbf{F}$, $\mathbf{F}^{\mathrm{ex}}$, and $\mathbf{G}$ is of course much less than the 81 each that one might imagine from the number of indices. First, since we are concerned only with strains, not rotations of the entire crystal, the strain tensor is symmetric, with but six independent elements, so that $\mathbf{F}$, $\mathbf{F}^{\mathrm{ex}}$, and $\mathbf{G}$ could have at most 54 elements each. Further reductions are

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[9] (a) C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1956), p. 85 ff; (b) In common parlance saturation magnetostriction depends on orientation of applied field, forced magnetostriction upon its magnitude. The usual mechanism for forced magnetostriction is indirect; by increasing $\langle S\rangle$ the applied field raises the contribution due to the F^{ex}- and G-type terms. On the other hand, the role of H in the F term is direct.

[10] J. Kondo, Progr. Theoret. Phys. (Kyoto) **28**, 1026 (1962); H. Watanabe, *ibid.* **18**, 405 (1957).

[11] B. C. Wybourne, Phys. Rev. **148**, 317 (1966).

[12] (a) T. G. Phillips and R. L. White, Phys. Rev. **153**, 616 (1967); (b) M. Blume, S. Geschwind, and Y. Yafet (to be published).

effected by considerations of the meaning of fictitious spin, and then of crystal symmetry.

A useful attribute of fictitious spin is its fictitiousness. The axes of quantization of this spin need not bear any simple relation to the crystal coordinates, but derive rather from the Hilbert space of the Hamiltonian.[12b] Being arbitrary up to a unitary transformation, they can be fixed by imposing conditions on three elements in $\mathbf{g}$, $\mathbf{g}^{\mathrm{ex}}$, and $\mathbf{D}$, or, in the strained crystal, on

$$\mathbf{g}+\mathsf{F}:\boldsymbol{\varepsilon};\quad \mathbf{g}^{\mathrm{ex}}+\mathsf{F}^{\mathrm{ex}}:\boldsymbol{\varepsilon};\quad \mathbf{D}+\mathsf{G}:\boldsymbol{\varepsilon}. \tag{2.6}$$

The choice of quantization axes of $\mathbf{S}$ is made on physical grounds. Of the three tensors, strained or not, the one most immediately measurable is $\mathbf{g}$ because it is coupled to the external field. The three Euler angles of the fictitious-spin coordinates are then conventionally fixed by the physically attractive specification that $[\mathbf{g}+\mathsf{F}:\boldsymbol{\varepsilon}]$ be symmetric, for then spin and moment will align for three orthogonal principal directions. Note that this does not imply that $\mathbf{S}$ is quantized along $\mathbf{H}$ for all directions, nor is any physical symmetry implied by a choice of spin axes.

Having exhausted our three degrees of freedom in specifying that total $\mathbf{g}$ be symmetric, we are not at liberty to impose additional constraints on $\mathbf{g}^{\mathrm{ex}}$. Thus, $\mathbf{g}^{\mathrm{ex}}$ has nine elements, the three antisymmetric terms being the Dzyaloshinski[13] interaction, not *a priori* small. On the other hand, because $\mathbf{D}$ is a single-ion term, and antisymmetric combinations of components of a single spin can be reduced by the commutation relations, $\mathbf{D}$ is perforce symmetric, provided time reversal invariance has been incorporated in the usual way. Furthermore, $\mathbf{D}$ has been adjusted to describe the separations of levels of a multiplet, but not the absolute positions with respect to any other multiplet. Thus, the trace of $\mathbf{D}$, which determines the center of gravity of the multiplet and which cannot be ascertained in an EPR experiment, can be set equal to zero. No similarly general argument can be made for the trace of $\mathbf{g}$ or $\mathbf{g}^{\mathrm{ex}}$, although in special cases changes in these traces may not contribute to angular-dependent quantities.[14]

Summing up these arguments, $\mathbf{g}$, $\mathbf{g}^{\mathrm{ex}}$, and $\mathbf{D}$ can have at most six, nine and five elements; and since $\boldsymbol{\varepsilon}$ has six, we expect that F, F^{ex}, and G can have at most 36, 54, and 30 elements.

So far we have said nothing of the effect of local site of symmetry, which of course will usually greatly reduce the number of magnetoelastic constants. How one imposes the restrictions of site symmetry is so well known that we do not repeat the arguments[15] here but present the forms of F, F^{ex}, and G for the 32 crystal-point groups. Dobrov[8] has already calculated G for all these groups. We follow the Voigt notation. The columns refer to strain components as defined by Love. The nine rows correspond to

$$\begin{gathered}\alpha_xS_x,\quad \alpha_yS_y,\quad \alpha_zS_z,\\ (\alpha_yS_z+\alpha_zS_y),\quad (\alpha_zS_x+\alpha_xS_z),\quad (\alpha_xS_y+\alpha_yS_x), \\ (\alpha_yS_z-\alpha_zS_y),\quad (\alpha_zS_x-\alpha_xS_z),\quad \text{and}\quad (\alpha_xS_y-\alpha_yS_x),\end{gathered} \tag{2.7}$$

where α_i are direction cosines of the magnetization $\mathbf{M}$ [see Eq. (2.4)].

For the 32 point groups, F^{ex} is given as follows (we drop the superscript on the F_{ij}^{ex} for clarity):

Triclinic System; Point Groups 1, $\bar{1}$:

$$(54 \text{ distinct, nonzero elements of } \mathsf{F}^{\mathrm{ex}}). \tag{2.8}$$

Monoclinic System; Point Groups 2, m, 2/m:

$$\mathsf{F}^{\mathrm{ex}}=\left[\begin{array}{cccccc} F_{11} & F_{12} & F_{13} & 0 & 0 & F_{16}\\ F_{21} & F_{22} & F_{23} & 0 & 0 & F_{26}\\ -(F_{11}+F_{21})+F_\alpha & -(F_{12}+F_{22})+F_\beta & -(F_{13}+F_{23})+F_\gamma & 0 & 0 & -(F_{16}+F_{26})+F_\delta\\ 0 & 0 & 0 & F_{44} & F_{45} & 0\\ 0 & 0 & 0 & F_{54} & F_{55} & 0\\ F_{61} & F_{62} & F_{63} & 0 & 0 & F_{66}\\ 0 & 0 & 0 & F_{74} & F_{75} & 0\\ 0 & 0 & 0 & F_{84} & F_{85} & 0\\ F_{91} & F_{92} & F_{93} & 0 & 0 & F_{96} \end{array}\right]. \tag{2.9}$$

Orthorhombic System; 222, mm2, mmm: F^{ex} is obtained from Eq. (2.9) by setting

$$F_{16}=F_{26}=F_{63}=F_{45}=F_{54}=F_{61}=F_{62}=F_\delta=F_{75}=F_{84}=F_{91}=F_{92}=F_{93}=0. \tag{2.10}$$

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[15] H. Callen, Am. J. Phys. **36**,735 (1968); E. Callen and H. Callen, Phys. Rev. **139**, A455 (1965).

Trigonal; *3*, $\bar{3}$:

$$\mathbf{F}^{\mathrm{ex}}=\begin{bmatrix} F_1+F_\alpha & F_2+F_\alpha & \frac{1}{2}(-F_3)+F_\gamma & F_{14} & -F_{25} & F_{16} \\ F_2+F_\alpha & F_1+F_\alpha & \frac{1}{2}(-F_3)+F_\gamma & -F_{14} & F_{25} & -F_{16} \\ -(F_1+F_2)+F_\alpha & -(F_1+F_2)+F_\alpha & F_3+F_\gamma & 0 & 0 & 0 \\ F_{41} & -F_{41} & 0 & F_{44} & F_{45} & F_{46} \\ -F_{46} & F_{46} & 0 & -F_{45} & F_{44} & F_{41} \\ -F_{16} & F_{16} & 0 & F_{25} & F_{14} & \frac{1}{2}(F_1-F_2) \\ F_{71} & -F_{71} & 0 & F_{74} & F_{75} & F_{76} \\ -F_{76} & F_{76} & 0 & -F_{75} & F_{74} & F_{71} \\ +F_{91} & F_{91} & F_{92} & 0 & 0 & 0 \end{bmatrix}. \qquad (2.11)$$

Trigonal; *32*, *3m* $\bar{3}m$: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.11) by setting

$$F_{16}=F_{25}=F_{45}=F_{46}=F_{75}=E_{76}=F_{91}=F_{92}=0. \qquad (2.12)$$

Tetragonal; *422*, *4/mmm*, *4mm*, $\bar{4}2m$: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.11) by setting

$$F_{14}=F_{16}=F_{25}=F_{41}=F_{45}=F_{46}=F_{71}=F_{75}=F_{76}=F_{91}=F_{92}=0, \qquad (2.13a)$$

and substituting

$$F_{66} \text{ for } \tfrac{1}{2}(F_1-F_2) \qquad (2.13b)$$

in the (6,6) position only.

Tetragonal; *4*, $\bar{4}$, *4/m*: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.11) by setting

$$F_{14}=F_{25}=F_{41}=F_{46}=F_{71}=F_{76}=0, \qquad (2.14a)$$

and substituting F_{66} for $\frac{1}{2}(F_1-F_2)$ in (6,6),

$$\begin{aligned} F_{61} &\text{ for } -F_{16} \text{ in } (6,1), \\ -F_{61} &\text{ for } F_{16} \text{ in } (6,2). \end{aligned} \qquad (2.14b)$$

Hexagonal; *6*, $\bar{6}$, *6/m*: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.11) by setting

$$F_{14}=F_{25}=F_{41}=F_{46}=F_{71}=F_{76}=0. \qquad (2.15)$$

Hexagonal; *622*, *6mm*, $\bar{6}m2$, *6/mmm*: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.10) by setting

$$F_{14}=F_{16}=F_{25}=F_{41}=F_{45}=F_{46}=F_{71}=F_{75}=F_{76}=F_{91}=F_{92}=0. \qquad (2.16)$$

Cubic; *23*, *m3*:

$$\mathbf{F}^{\mathrm{ex}}=\begin{bmatrix} F_1+F_\alpha & F_2+F_\alpha & -(F_1+F_2)+F_\alpha & 0 & 0 & 0 \\ -(F_1+F_2)+F_\alpha & F_1+F_\alpha & F_2+F_\alpha & 0 & 0 & 0 \\ F_2+F_\alpha & -(F_1+F_2)+F_\alpha & F_1+F_\alpha & 0 & 0 & 0 \\ 0 & 0 & 0 & F_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & F_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & F_{44} \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}. \qquad (2.17)$$

Cubic; *432*, $\bar{4}3m$, *m3m*: $\mathbf{F}^{\mathrm{ex}}$ is obtained from Eq. (2.17) by substituting

$$-\tfrac{1}{2}F_1 \text{ for } F_2. \qquad (2.18)$$

Equations (2.8) to (2.18) describe the exchange magnetoelastic tensor. To obtain $\mathbf{F}$, the strain derivative of the paramagnetic g matrix, one sets all antisymmetric elements, those in rows seven, eight, and nine, equal to zero. The traces of the matrices $\mathbf{g}^{\mathrm{ex}}$ and $\mathbf{g}$ are not necessarily strain invariant, that is

$$\sum_{i=1}^{3} F_{ik} \neq 0, \quad \sum_{i=1}^{3} F_{ik}^{\mathrm{ex}} \neq 0, \quad k=1,6 \qquad (2.19)$$

and to the extent that $\langle\mathbf{S}\rangle$ is not parallel to $\mathbf{H}$ or $\mathbf{M}$, these traces retain physical significance. In contrast, $\mathbf{D}$ and its strain derivative, $\mathbf{G}$ are traceless. To obtain $\mathbf{G}$ for all point groups, set all elements in rows seven, eight, and nine equal to zero and everywhere set

$$F_\alpha=F_\beta=F_\gamma=F_\delta=0. \qquad (2.20)$$

The representations of $\mathbf{G}$ obtained by these reductions agree with Dobrov.[8]

B. Magnetoelastic Energy

Magnetostriction is a gross distortion, adequately described by the (strain-dependent) expectation value of $\mathcal{H}^{\mathrm{me}}$. This expectation value is called magnetoelastic energy, since it is linear in the strain and the elastic energy quadratic, minimum free energy is attained at some nonzero strain. Taking the expectation value involves averaging the spin operators in the density matrix of the unperturbed spin Hamiltonian. The strain operators are treated classically.

When crystal-field terms such as $\mathbf{S}\cdot\mathbf{D}\cdot\mathbf{S}$ are comparable to the paramagnetic Zeeman and exchange terms of Eq. (2.1), the resultant magnetoelastic energy is somewhat complicated; we defer discussion of this circumstance to Sec. IV. The more simple and common case, the one we discuss now, occurs when the Zeeman terms dominate. We may then define a spin direction when there is rotation symmetry about the quantization axis. Denoting by α_i, $i=1, 2, 3$, the direction cosines of the spin-quantization axis with respect to the crystal axes and by $\langle S_\zeta\rangle$ the quantum-mechanical average of the fictitious spin along this axis, one finds for the magnetoelastic energy

$$\mathcal{E}^{\mathrm{me}}=\langle\mathcal{H}^{\mathrm{me}}\rangle=\sum_{ijkl}^{3}\{H_i\alpha_j\langle S_\zeta\rangle F_{ijkl}+M_i\alpha_j\langle S_\zeta\rangle F_{ijkl}{}^{\mathrm{ex}}+\tfrac{1}{2}\alpha_i\alpha_j[3\langle S_\zeta{}^2\rangle-S(S+1)]G_{ijkl}\}\epsilon_{kl}. \quad (2.21)$$

In some materials, the paramagnetic and exchange g factors are isotropic. The spin axis and magnetization then coincide with the external field. The magnetoelastic energy then simplifies to

$$\mathcal{E}^{\mathrm{me}}=\alpha_i\alpha_jB_{ijkl}\epsilon_{kl}, \quad (2.22)$$

where

$$B_{ijkl}=|H|\langle S_\zeta\rangle F_{ijkl}+M\langle S_\zeta\rangle F_{ijkl}{}^{\mathrm{ex}}+\tfrac{1}{2}[3\langle S_\zeta{}^2\rangle-S(S+1)]G_{ijkl} \quad (2.23)$$

and α_i is the direction cosine of the applied field. When either g or $\mathbf{g}^{\mathrm{ex}}$ is anisotropic, Eq. (2.22) for $\mathcal{E}^{\mathrm{me}}$ does not apply, and Eq. (2.21) must be used in its full form. A case in point is Yb^{3+} in the iron garnet, to which we now turn.

III. Yb^{3+} IN GARNET

A. Magnetization

Yb^{3+} in the garnets is a much studied system and in many regards a well understood one.[16] The crystal field splits the lowest $J=\frac{7}{2}$ multiplet into Kramers doublets, the lowest of which is more than 500 cm^{-1} below the nearest excited state.[17] Since an effective spin $S=\frac{1}{2}$ cannot support a crystal-field term, the behavior of the ground doublet is summarized by a spin Hamiltonian of the form

$$\mathcal{H}=+\mathbf{H}\cdot\mathbf{g}\cdot\mathbf{S}+\mathbf{M}_{\mathrm{Fe}}\cdot\mathbf{g}^{\mathrm{ex}}\cdot\mathbf{S}, \quad (3.1)$$

with $\mathbf{M}_{\mathrm{Fe}}(T)$ the net temperature-dependent moment of the two-iron sublattices. Both the paramagnetic and exchange g tensors have been shown to be very anisotropic.[18]

The garnet unit cell contains 12 rare-earth (c) sites, six inequivalent ones need be considered. At each site there is approximate orthorhombic symmetry (222) with principal axes obtained by rotating about the x, y or z cubic axis by ±45°. Dillon and Walker[19] illustrate the structure. Besides the rare-earth sites, there are a and d sites occupied by Fe^{3+} in the ferrimagnetic garnet. The rare earths align themselves in the exchange field of the iron (J_{cd}), but their responsiveness is largely unrequited for the irons care mostly for each other,[20] $J_{ad}\gg J_{cd}\gg J_{cc}$. We may envision the iron sublattices as locked to form a net iron moment, positioned by the external field with a correction for anisotropy. The rare earths on each site, however, will seek their own equilibria.

We will assume that the iron moment is along the axis of the external field. This is never really true except for symmetry directions, and even then, near the compensation temperature (7.6°K) peculiar things may occur.[21] Specifically in the neighborhood of 7.6°K, magnetic fields of less than 10 kOe will cause the rare earth and iron sublattices to cant away from the external-field direction. At 13°K, the dominant iron moment lies along the field and the rare earth is antiparallel, up to a field of 35 kOe, above which field strength the iron begins to swing away and the rare earth rotates up toward the field direction. Throughout the present paper we shall ignore such effects, assuming that the external-field strength is sufficient to eliminate domains, align the net moment along the field direction but not so strong as to cause canting of sublattice moments.

The physics of the Hamiltonian, Eq. (3.1), is then summarized[22] by a spin-quantization direction $\hat{s}$ and an energy splitting Δ:

$$\Delta=|\mathbf{H}\cdot\mathbf{g}+\mathbf{M}_{\mathrm{Fe}}\cdot\mathbf{g}^{\mathrm{ex}}|, \quad (3.2)$$

$$\hat{s}=\frac{\mathbf{H}\cdot\mathbf{g}+\mathbf{M}_{\mathrm{Fe}}\cdot\mathbf{g}^{\mathrm{ex}}}{\Delta}. \quad (3.3)$$

The moment of a rare-earth ion on site σ is then

$$\mathbf{M}^\sigma=\mu_\beta\langle\mathbf{S}^\sigma\rangle\cdot\mathbf{g}, \quad (3.4)$$

where

$$\langle\mathbf{S}^\sigma\rangle=\frac{\hat{s}}{2}\tanh\frac{\Delta^\sigma}{2k_BT}. \quad (3.5)$$

The ith component of the total rare earth moment at

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temperature T and in the limit of zero field is

$$M_{\mathrm{RE}\,i}=\sum_{\sigma=1}^{6} M_i{}^{\sigma}=\mu_\beta \sum_{\sigma=1}^{6} \tfrac{1}{2}\tanh\frac{\Delta^\sigma}{2k_BT} \times \sum_{kj} \frac{(M_{\mathrm{Fe}})_k g_{kj}{}^{\mathrm{ex}} g_{ji}}{\Delta^\sigma}. \quad (3.6)$$

Henderson and White[23] carried out this sort of calculation of the moment along with that of other properties of Yb^{3+} in YIG. However, they erroneously applied a formula meant for a paramagnet in a real, rather than an exchange, field. The correct result from Eq. (3.6) at $T=0°\mathrm{K}$ is

$$M_{\mathrm{RE}[100]}=10.22\ \mu_B(\text{six Yb's, field along }[100]),$$

$$M_{\mathrm{RE}[111]}=10.07\ \mu_B(\text{six Yb's, field along }[111]),$$

and the anisotropy in the moment is opposite to that calculated in Ref. 23. In any event, the small anisotropy has not been measured.[24]

B. Magnetoelastic Energy for Yb^{3+}

Since there are no crystal-field terms in the spin Hamiltonian of a Kramer's doublet, the magneto-elastic Hamiltonian, Eq. (2.4), reduces for Yb^{2+} in the garnet to

$$\mathcal{H}^{\mathrm{me}\,\sigma}=(\mathbf{H}\cdot\mathbf{F}^{\sigma}\cdot\mathbf{S}+\mathbf{M}_{\mathrm{Fe}}\cdot\mathbf{F}^{\mathrm{ex}\sigma}\cdot\mathbf{S}):\boldsymbol{\varepsilon}. \quad (3.7)$$

The two $\mathbf{F}$ tensors bear a site index σ because, although the $\mathbf{F}$ tensor looks the same in the local site coordinate systems, the tensor must be transformed by the unitary transformation which carries the local orthorhombic site axes into the macroscopic cubic coordinates. It is this rotated representation which is implied by $\mathbf{F}^\sigma$. To find the magnetoelastic energy (of site σ) at zero external field we then need only calculate the expectation value of the $\mathbf{F}^{\mathrm{ex}}$ term in Eq. (3.7), using the result of Eqs. (3.4) and (3.5):

$$\mathcal{E}^{\mathrm{me}\,\sigma}(H=0)=\left(\mathbf{M}_{\mathrm{Fe}}\cdot\mathbf{F}^{\mathrm{ex}\,\sigma}\cdot\frac{\hat{s}}{2}\tanh\frac{\Delta^\sigma}{2k_BT}\right):\boldsymbol{\varepsilon}. \quad (3.8)$$

This can be expressed more conveniently in terms of the magnitude $\mathbf{M}_{\mathrm{Fe}}$ and direction cosines α_i of the iron magnetization $\mathbf{M}_{\mathrm{Fe}}$ by means of Eqs. (3.2) and (3.3):

$$\mathcal{E}^{me}=\sum_{\sigma} \tfrac{1}{2}M_{\mathrm{Fe}}\tanh\frac{\Delta^\sigma(\alpha)}{2k_BT} \times \sum_{ijklm=1}^{3} \frac{\alpha_i\alpha_m g_{mj}{}^{\mathrm{ex}\ \sigma}F_{ijkl}\epsilon_{kl}}{\Delta^\sigma(\alpha)}, \quad (3.9)$$

where

$$\Delta^\sigma(\alpha)=M_{\mathrm{Fe}}\Big[\sum_{ij}(\alpha_i g_{ij}{}^{\mathrm{ex}\ \sigma})^2\Big]^{1/2}. \quad (3.10)$$

The dependence of the exchange splitting of the σth site on the direction of $\mathbf{M}_{\mathrm{Fe}}$ is emphasized by the notation $\Delta^\sigma(\alpha)$. The site dependence of the splitting enters Eq. (3.10) through the representation of the $\mathbf{g}^{\mathrm{ex}}$ tensor of site σ in the macroscopic coordinate system. We note that because of the anisotropy of $\mathbf{g}^{\mathrm{ex}}$, $\mathcal{E}^{\mathrm{me}\,\sigma}$ is not second order in α, but involves all orders.

C. Comparison with Phillips and White

Phillips and White (PW) have already given a spin-Hamiltonian analysis of the magnetostriction of rare-earth ions in garnets and of iron group monoxides,[5,12,25] so that our analysis must properly be related to theirs. PW discuss three rare-earth substitutions in YIG[5]—Gd^{3+}, Yb^{3+} and Er^{3+}—the first because it is an S-state ion, and Yb^{3+} and Er^{3+} for which the ground states were presumed to be Kramer's doublets. PW conjectured the magnetostriction of Gd^{3+} to be due to the (single-ion) G term of Eq. (2.4). Later experiments (Ref. 11) indicate that the source is the (two-ion) $\mathbf{F}^{\mathrm{ex}}$ term. As for Er^{3+}, spectroscopic evidence[18,26] suggests that the exchange splittings are in fact comparable to the crystal-field separations. The third-ion PW consideration is Yb^{3+}.

They relate the magnetostriction constants of the exchange-coupled YbIG to their measurements of the strain dependence of the paramagnetic g tensor of YbGaG.

We recall that [Eqs. (2.1) to (2.3)]

$$\mathbf{g}^{\mathrm{ex}}=\boldsymbol{\lambda}\cdot\mathbf{g}, \quad (3.11)$$

where λM defined an exchange field whose effect on the paramagnetically situated ion would duplicate the field due to exchange in the iron garnet. Now differentiating,

$$\mathbf{F}^{\mathrm{ex}}=\frac{\partial\mathbf{g}^{\mathrm{ex}}}{\partial\epsilon}=\frac{\partial\lambda}{\partial\epsilon}\cdot\mathbf{g}+\boldsymbol{\lambda}\cdot\frac{\partial\mathbf{g}}{\partial\epsilon}=\frac{\partial\lambda}{\partial\epsilon}\cdot\mathbf{g}+\boldsymbol{\lambda}\cdot\mathbf{F}. \quad (3.12)$$

The bifurcation in Eq. (3.12) is more than formal emphasizing a distinction between the effect of strain on the exchange interaction itself (the first term) and the strain dependence of the responsiveness of the ion to effective fields. The first term is not measured in

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[23] J. W. Henderson and R. L. White, Phys. Rev. **123**, 1627 (1961).

[24] F. W. Harrison, J. F. A. Thompson, and K. Tweedale [in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (Institute of Physics and The Physical Society, London, 1965), p. 664], performed the measurements and also point out the error in Ref. 23.

[25] R. L. White and T. G. Phillips, J. Appl. Phys. **39**, 579 (1968); Phys. Rev. Letters **16**, 650 (1966); J. Appl. Phys. **38**, 1222 (1967).

[26] S. Hufner, P. Kienle, W. Wiedmann, J. Frey, and W. Zinn, in *Proceedings of the International Conference on Magnetism, Nottingham, 1965* (Institute of Physics and The Physical Society, London, 1965), p. 672; S. Hufner and H. Schmidt, Physik Kondensierten Materie **4**, 262 (1965).

EPR on paramagnetic garnets, and awaiting results in the iron garnet,[27] we turn to the second term.

One then finds

$$\mathcal{E}^{\mathrm{me}}=\sum_{\sigma} \mathbf{M}_{\mathrm{Fe}}\cdot\boldsymbol{\lambda}^{\sigma}\cdot\mathbf{F}^{\sigma}\cdot\langle\mathbf{S}^{\sigma}\rangle. \tag{3.13}$$

The effective-exchange field on the rare-earth ion, $H^{\mathrm{ex}\sigma}=\mathbf{M}_{\mathrm{Fe}}\cdot\boldsymbol{\lambda}^{\sigma}$, has been measured by Wickersheim[18]; its principal values in the local orthorhombic coordinate system are

$$\begin{aligned} H_x^{\mathrm{ex}}&=87.2\ \mathrm{kOe},\\ H_y^{\mathrm{ex}}&=153.9\ \mathrm{kOe},\\ H_z^{\mathrm{ex}}&=169.0\ \mathrm{kOe}. \end{aligned} \tag{3.14}$$

The change in part of the magnetoelastic energy with strain, and hence that part of the magnetostriction which is due to the second term in Eq. (3.12), should be found from Eq. (3.13) after having determined the elements of **F**.

PW report measurements of many of the components of F but unfortunately the experimental configuration is such that pressure cannot be applied parallel to the field direction as the magnet gets in the way. Thus three of the 12 components of **F** could not be measured. To complete the **F** tensor PW invoke an assumption of "tracelessness."

$$\sum_{i=1}^{3} F_{iikl}=0, \quad k,l=1,2,3 \tag{3.15}$$

or, in the notation of Eqs. (2.10),

$$F_{\alpha}=F_{\beta}=F_{\alpha}=0. \tag{3.16}$$

PW did not intend their argument to be rigorous (and in a point-charge calculation demonstrate it to be only approximate). Actually, the errors in the matrix elements introduced by this assumption can be of the same size as the elements themselves. Furthermore, we shall see that changes in Trg do contribute to angular terms in the magnetoelastic energy. On the other hand, PW have correctly noted that it is not necessary to measure all 12 orthorhombic elements of F in order to determine the contribution to the two parameters in the cubic magnetoelastic energy, when the latter is only second order in direction cos of the field. In fact, PW apparently used such an abbreviated approach in computing the magnetostriction constants.[28]

There is another matter which deserves note. PW write that the change of energy of a paramagnetic transition in YbGaG is

$$\delta E=H\delta g_{ij}\alpha_i\alpha_j, \tag{3.17}$$

where α_i are the direction cosines of H and δg is the change of g with strain. The more correct relation, applicable when g is anisotropic, is obtained by differentiating Δ [see Eq. (3.2)]:

$$\delta E=H\frac{\delta g_{ij}g_{kj}\alpha_i\alpha_j}{[\sum_i(g_{ij}\alpha_j)^2]^{1/2}}. \tag{3.18}$$

There is no distinction between Eqs. (3.17) and (3.18) for principal directions, but it would seem that these were not always accessible. For aribtrary directions, very considerably different values for δg might be obtained, since the principal values of g itself differ by 35%.

D. Magnetostriction of Yb^{3+} in YIG

For a cubic material such as YbIG, the magnetostriction may be expanded as

$$\begin{aligned}\frac{\Delta l}{l}=&\tfrac{3}{2}\lambda_{100}(\alpha_1^2\beta_1^2+\alpha_2^2\beta_2^2+\alpha_3^2\beta_3^2-\tfrac{1}{3})\\ &+3\lambda_{111}(\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_1\alpha_3\beta_1\beta_3)+\lambda^{\gamma 4}\{\tfrac{1}{2}[\alpha_3^4\\ &-\tfrac{1}{2}(\alpha_2^4+\alpha_1^4)-\tfrac{9}{7}(\alpha_3^2-\tfrac{1}{3})][\beta_3^2-\tfrac{1}{3}]+\tfrac{1}{4}[\alpha_1^4-\alpha_2^4\\ &-\tfrac{6}{7}(\alpha_1^2-\alpha_2^2)][\beta_1^2-\beta_2^2]\}+\lambda^{\epsilon,4}\{\alpha_1\alpha_2(\alpha_3^2-\tfrac{1}{7})\\ &\times\beta_1\beta_2+\alpha_1\alpha_3(\alpha_2^2-\tfrac{1}{7})\beta_1\beta_3+\alpha_2\alpha_3(\alpha_1^2-\tfrac{1}{7})\beta_2\beta_3\}\\ &+\lambda^{\alpha,4}\{\alpha_1^4+\alpha_2^4+\alpha_3^4-\tfrac{3}{5}\}+O(\alpha^6).\end{aligned} \tag{3.19}$$

$\Delta l/l$ is fractional change in length of a strain gauge, α_i give the moment direction of the iron sublattice, β_i are the direction cosines along which the strain gauge is placed, and the λ's are the magnetostriction constants. λ_{100} and λ_{111} are written in the traditional form.[1]

It is awkward to obtain analytically the coefficients in Eq. (3.19) from the magnetoelastic energy, Eq. (3.9). A far easier task is to obtain $\Delta l/l$ for specific values of α_i and β_i. We have, therefore, proceeded with the calculation by computing $\Delta l/l$ for sets of directions and fitting the results to a truncated form of Eq. (3.19). For example, if we assume that second-order terms along suffice, we may write

$$\begin{aligned}\lambda_{100}&=\frac{2}{3}\left(\frac{\Delta l}{l}\bigg|_{\beta=100}^{\alpha=100}-\frac{\Delta l}{l}\bigg|_{\beta=010}^{\alpha=100}\right),\\ \lambda_{111}&=\frac{3}{4}\left(\frac{\Delta l}{l}\bigg|_{\beta=111}^{\alpha=111}-\frac{\Delta l}{l}\bigg|_{\beta=11-1}^{\alpha=111}\right).\end{aligned} \tag{3.20}$$

If we employ the F elements given by PW,[5] the $\mathbf{g}$ and $\mathbf{H}_{\mathrm{eff}}$ of Wickersheim,[18] and the stiffness constants Clark and Strakna,[29] then Eq. (3.20) yields the following

---

[27] Total F^{ex} would be measured by exchange splittings in the *iron* garnet under pressure. However, in YbIG, the relevant absorptions are obscured by a strong iron absorption and only thin samples, difficult to strain, have been studied. Comstock, Buchanan, and White (Ref. 7) have measured the combinations of components of F^{ex} which give the cubic magneostriction associated with the $^2F_7$ state of Ce^{3+} in YIG. They scale these and relate them to magnetostriction in Yb-doped YIG (where $^2F_{7/2}$ is indeed the ground state).

[28] T. G. Phillips (private communication).

[29] A. E. Clark and R. E. Strakna, J. Appl. Phys. **32**, 1172 (1961).

contributions to the λ's at 0°K:

$$\lambda_{100}=185\times10^{-6},\quad \lambda_{111}=36.5\times10^{-6}. \tag{3.21}$$

However, one recognizes that the anisotropic $\mathbf{g}$ and $\mathbf{H}_{\text{eff}}$ cause not only second-, but also fourth-, sixth-, and higher-degree magnetostriction terms in Eq. (3.19), and $\Delta l/l$ must be evaluated accordingly. Truncating at fourth-degree terms, and evaluating in an appropriate set of strain and field directions, we compute at 0°K

$$\lambda_{100}=199\times10^{-6},\quad \lambda_{111}=44\times10^{-6}, \tag{3.22}$$

so the error in using the simpler Eq. (3.20) is not great even at $T=0$°K. PW employ the same data, but conclude that these imply

$$\lambda_{100}=82\times10^{-6},\quad \lambda_{111}=34\times10^{-6}. \tag{3.23}$$

The experimental values for low temperatures are[30] $\lambda_{100}=49\times10^{-6}$, $\lambda_{111}=-27\times10^{-6}$, and[31] $\lambda_{100}=97\times10^{-6}$, $\lambda_{111}=-31\times10^{-6}$. There are also other, higher-temperature measurements.[32a] Generally, results are extrapolated from 10% doped samples, which may explain some of the experimental scatter. In any event, these do not corroborate the theory, but then it should be recalled we (along with PW) have only tried to get one of the two contributions in Eq. (3.12). The thought here has been to illustrate the structure of the theory.

IV. TEMPERATURE DEPENDENCE

A. Introduction

The usual treatment of the statistical mechanics of magnetostriction originated by Kittel and Van Vleck,[3] parallels that of magnetic anisotropy and reduces to the same $\frac{1}{2}l(l+1)$ power law at low temperatures, ignoring the effect of crystal fields. Ultimately, the justification for the treatment requires (a) that the thermal probability of an individual spin being in state i with quantum average component of magnetic moment $\langle\mu_z\rangle_i$ be

$$P_i\propto e^{X\langle\mu_z\rangle_i}, \tag{4.1}$$

and (b) that the $\langle\mu_z\rangle_i$ be proportioned to successive integers

$$\langle\mu_z\rangle_i=am_i. \tag{4.2}$$

Requirement (a) has been demonstrated to be satisfied on various statistical models.[32b] The quantity X, which is a function of temperature, is entirely arbitrary.

When the individual states are Zeeman states, (b) follows. As a result,[2] single-ion magnetoelastic terms of degree l produce (for infinite spin) a temperature dependence $\hat{I}_{l+\frac{1}{2}}[\mathcal{L}^{-1}(m)]$, where $\hat{I}_{l+\frac{1}{2}}$ is a reduced hyperbolic Bessel function, and $\mathcal{L}^{-1}$ is the inverse Langevin function of the reduced sublattice magnetization m. Thus, the contribution of a sublattice to λ_{100} and λ_{111}, the $l=2$ magnetostriction coefficients of cubic crystals, should vary as $\hat{I}_{5/2}$. For finite spin, the treatment is not greatly different.

This was the case for a single-ion source term such as $\mathbf{G}$. The two-ion term $\mathbf{F}^{\text{ex}}$ produces $l=2$ magnetostriction coefficients which are sufficiently approximated by m^2, or rather M_AM_B if the magnetoelastic term couples spins on sublattices A and B.

This treatment has been adequate, in fact immoderately successful, in describing the magnetostrictive behavior of metals—nickel,[33] dysprosium,[34] terbium[35]—where electron shielding mostly eliminates the crystal field, and of S-state ions in garnets[36,37] and spinels.[38] But for degenerate ground-state ions in nonconductors, such as most rare earths in garnets, one expects more complex behavior. These complexities can arise both from anisotropic exchange and from relatively large crystal fields. As an example of the effect of anisotropic exchange in a Kramers ion, we again turn to YbIG, and elaborate on the implications of Eqs. (2.21) and (3.9). To display the crystal-field effect in a non-Kramers ion, we study a model related to DyIG.

B. Kramers Ion: Yb^{3+} in YIG

As we have mentioned in Sec. III, since only $\mathbf{F}$ and $\mathbf{F}^{\text{ex}}$ terms can contribute to the magnetostriction of Yb^{3+} in YbIG, the simplest statistical theory[2] would suggest that the strains vary as $M_{\text{RE}}(T)$ in the former case and $M_{\text{RE}}(T)M_{\text{Fe}}(T)$ in the latter. Of these two terms, the direct forced magnetostriction term ($\mathbf{F}$ term) has not been studied, although sufficiently high fields are now available.

Let us confine attention to the intrinsic magnetostriction ($\mathbf{F}^{\text{ex}}$ term), and compare the implications of Eq. (3.9) with the conventional result. Rigorous analysis is complicated by the occurrence in Eq. (3.19) of polynomials of high order in the direction cosines of the magnetization, particularly at low temperatures [see Eqs. (3.21) and (3.22)].

The result of the more simple Eq. (3.20), of which (3.21) is the $T=0$°K limit, is obtainable in closed

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[32] (a) S. Iida, J. Phys. Soc. Japan **22**, 1201 (1967); N. Bertram and R. V. Jones, J. Appl. Phys. **38**, 122S (1967); (b) For a review of the statistical mechanics of magnetic anisotropy see H. B. Callen and Earl Callen, J. Phys. Chem. Solids **27**, 1271 (1966); H. B. Callen and S. Shtrikman, Solid State Commun. **3**, 5 (1965).

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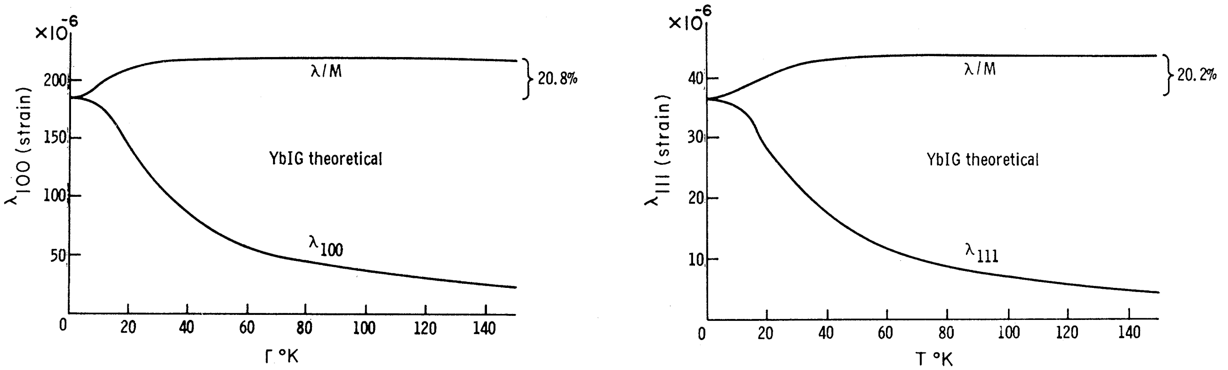

(a) (b)

FIG. 1. λ versus temperature for the strain-dependent *g*-factor term in YbIG. Were it not for averaging over sublattices, λ/M (the ratio of second-order magnetostriction to magnetization) would be constant. As it is, constancy is maintained except at the lowest temperatures.

form:

$$
\begin{aligned}
10^6\lambda_{100} &= 188 \tanh\frac{40.1}{T} - 3.5 \tanh\frac{16.7}{T}\,, \\
10^6\lambda_{111} &= 74.1 \tanh\frac{36.4}{T} - 37.6 \tanh\frac{31.7}{T}\,,
\end{aligned}
\tag{4.3}
$$

where T is in °K. The temperature factors are associated with the splittings when the field is along [100] and [111] for λ_{100} and λ_{111}, respectively. Specifically, for [100] there are two sites with 16.7°K splittings and four split by 40.1°K. For [111] there are three of each kind.

Next let us calculate the moment. Using the same data as above, the magnetizations[24] in the two principal directions are

$$
\begin{aligned}
M_{100}(T) &= 7.37 \tanh\frac{40.1}{T} + 2.85 \tanh\frac{16.7}{T}\,, \\
M_{111}(T) &= 5.12 \tanh\frac{36.4}{T} + 4.95 \tanh\frac{31.7}{T}\,.
\end{aligned}
\tag{4.4}
$$

Although the magnetostriction contribution from each type of site has the same temperature dependence as does the moment contribution from that site, the total magnetostriction is not proportional to the total moment—the weighting of the two terms in λ_{100} (or in λ_{111}) in Eq. (4.3) is different from the weightings in Eq. (4.4). The results of this calculation are illustrated in Fig. 1. [We warn that we do not expect Eq. (4.3) to agree quantitatively with the observed temperature dependence of λ_{100} or λ_{111} of YbIG, since the weightings of the terms depend on the particular elements in the $\mathbf{F}^{\text{ex}}$ tensor.]

There are, nonetheless, a number of points which the calculation illustrates. One major distinction between the results of the isotropic and anisotropic exchange theories is that, with isotropic exchange, all the terms in the Hamiltonian which we have considered, in Eq. (2.4), can lend at most to second-degree magnetostriction, such as λ_{100} and λ_{111}. But with anisotropic exchange, the same source terms lead to magnetostriction coefficients of all even degrees. A second significant result concerns the relationship between magnetostriction and magnetization. In YbIG, because of the anisotropic exchange interaction, neither $\lambda_{100}(T)$ nor $\lambda_{111}(T)$ should be precisely proportional to $M_{\text{RE}}(T)$. At the same time, $\lambda_{100}(T)$ and $\lambda_{111}(T)$ surely vary more like $M_{\text{RE}}(T)$ than like $\hat{I}_{5/2}$, an inequality which Comstock and Raymond[31] employ to argue that the two-ion term is indeed the source of the Yb^{3+} magnetostriction in the garnet. Our result is, of course, for just part of the magnetostriction, but considering the role of averaging over sites, the above conclusion seems rather general, that is, λ is not strictly proportional to M, but it is not expected to deviate much.

Next we consider the contributions to λ_{100} and λ_{111} from the various elements in the local $\mathbf{F}^{\text{ex}}$. It will be recalled that in (222) symmetry, $\mathbf{F}^{\text{ex}}$ contains 15 elements including three Dzyaloshinski-type terms. When the six inequivalent garnet sites are summed over, we find no simple relation between the local symmetry of an element and its role in the cubic magnetostrictions. Specifically the Dzyaloshinski-type terms can contribute to both λ_{111} and λ_{100}. The model, dealing as it does with $\mathbf{F}$ rather than $\mathbf{F}^{\text{ex}}$, has artifically excluded these antisymmetric interactions. However, both as regards RE-Fe interaction symmetries[20] and RE site symmetry [see (2.10)] the antisymmetric terms should be present.

The final conclusion concerns the temperature dependence of the magnetostriction and magnetization at high temperatures. Figure 1 shows these both to go ultimately as $1/T$. This obtained by expanding tanh $(\Delta/2k_BT)$ for small arguments. We note that the second-degree magnetostrictions (λ_{100} and λ_{111}) of Eq. (3.19) go as $1/T$, fourth-degree as $1/T^3$, sixth-degree as $1/T^5$, and so on. Thus, as might be expected, the high-degree λ's enter strongly only at low temperatures.

C. Magnetostriction with Large Crystal Fields: Model of DyIG

The effect of strong-crystal fields on the behavior of magnetostriction is something about which it is difficult to theorize with generality. The magnetoelastic free energy at temperature T is to be found by taking the thermal expectation value of the perturbative terms of Eq. (2.4) over the crystal-field eigenstates. Whereas, the exchange-field eigenstates were Zeeman-like, and equally spaced, the crystal-field eigenstates are generally complex mixtures of Zeeman states, and are irregularly separated in energy. Still, if the unperturbed Hamiltonian and the tensors in Eq. (2.4) are known, the averaging can be carried out. Unfortunately $\mathbf{F}^{\text{ex}}$ and $\mathbf{G}$ for the Dy^{3+} ion, and every other, are almost entirely unknown. Consequently, barred both from a general orderly analysis and a particularized calculation, we have chosen to perform several model calculations whose purpose is to display some of the effect which can be expected.

The model in some sense relates to Dy^{3+} in the iron garnet. For this ion some data are available for the exchange field and for the crystal field, which is of comparable size. Although there is every reason to expect an exchange to be as anisotropic for the Dy^{3+} ion as for the Yb^{3+} ion in the garnet,[20,39a] we employ a scalar exchange, as only this number is available. This scalar exchange applies to the unperturbed J multiplet. Since crystal fields have very confusing effects on the response of an ion to exchange, it does not seem that any richness of effects is lost in this application.[39b] The value for H^{ex} is 327 kOe[40] $[2(g_J-1)=\frac{2}{3}]$.

As for crystal-field parameters, we use the A's calculated from ytterbium gallium garnet (YbGaG) data.[41] In the most naive theory these parzmetrize the c-site potential and do not depend on the ion occupying the site.[42] Better values for Dy may come from considering spectroscopic data[26,43] but would add little to our model. We have an unperturbed Hamiltonian including crystal fields of D_2 symmetry with an exchange field, and the magnitudes are reasonable for Dy^{3+} in garnets.

The effective spin for the lowest J multiplet in Dy^{3+} is 15/2. The symmetry of the Dy^{3+} site is orthorhombic. The most general form for $\mathcal{H}^{\text{me}}$ contains terms involving many spin operators related to the strains through tensor which themselves have many independent elements. We content ourselves with an examination of the following four models incorporating only symmetric terms of the form given in Eq. (2.4):

2-ion A model: $\mathcal{H}^{\text{me}\,\sigma}=\mathbf{M}_{\text{Fe}}\cdot\mathbf{F}\cdot\mathbf{J}\cdot\boldsymbol{\varepsilon}$,

$$\mathbf{F}=\begin{bmatrix} 1 & -\frac{1}{2} & -\frac{1}{2} & 0 & 0 & 0 \\ -\frac{1}{2} & 1 & -\frac{1}{2} & 0 & 0 & 0 \\ -\frac{1}{2} & -\frac{1}{2} & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}\times\frac{1}{J|M_{\text{Fe}}|},$$

where M_{Fe} stands for the magnetization of the iron sublattice (taken as temperature independent), the units of $\mathbf{F}$ are in cm^{-1}/strain, and the normalization is chosen so that the maximum contribution to $\mathcal{E}^{\text{me}}$ is of order 1 cm^{-1}/site ($J=15/2$). $\mathbf{F}$ is referred to the orthorhombic axes of the site σ. F is referred to the orthorhombic axes of the site σ. The model is 2-ion because the strain-dependent interaction involves both a Dy and an Fe.

1-ion A model: $\mathcal{H}^{\text{me}\,\sigma}=\mathbf{J}\cdot\mathbf{G}\cdot\mathbf{J}:\boldsymbol{\varepsilon}$,

$$\mathbf{G}=\begin{bmatrix} 1 & -\frac{1}{2} & -\frac{1}{2} & 0 & 0 & 0 \\ -\frac{1}{2} & 1 & -\frac{1}{2} & 0 & 0 & 0 \\ -\frac{1}{2} & -\frac{1}{2} & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}\times\frac{1}{J(J-\frac{1}{2})}.$$

Again, the normalization is such that $\mathcal{E}^{\text{me}}$ is of order 1 cm^{-1}/atom. This is 1-ion, since only the Dy is involved.

2-ion B model: $\mathcal{H}^{\text{me}\,\sigma}=\mathbf{M}_{\text{Fe}}\cdot\mathbf{F}\cdot\mathbf{J}:\boldsymbol{\varepsilon}$,

$$\mathbf{F}=\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix}\times\frac{1}{J|M|}.$$

1-ion B. model: $\mathcal{H}^{\text{me}\,\sigma}=\mathbf{J}\cdot\mathbf{G}\cdot\mathbf{J}:\boldsymbol{\varepsilon}$

$$\mathbf{G}=\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix}\times\frac{1}{J(J-\frac{1}{2})}.$$

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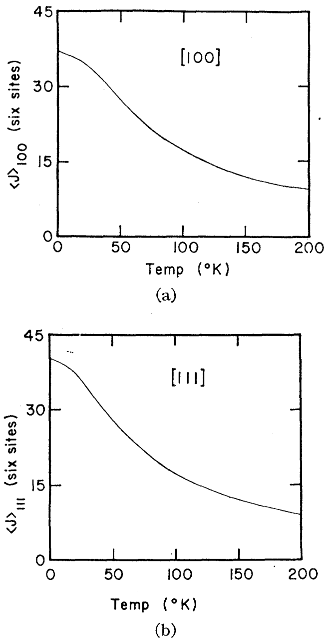

FIG. 2. $\langle J \rangle$ versus temperature for the DyIG model; (a) field along [100], (b) field along [111]. The iron lattices are taken as insensitive to temperature. Note the considerable anisotropy in $\langle J \rangle$ at low temperature: $\langle J \rangle_{111}$ is 10% larger than $\langle J \rangle_{100}$.

For each model we compute the cubic magnetostriction constants, λ_{100} and λ_{111} as defined by Eq. (3.20). This involves (numerically) getting the thermal expectation value of $\mathcal{H}^{\mathrm{me}\,\sigma}$ in the crystal and molecular-field eigenstates for each site σ and for selected directions of the iron magnetization. The effect of crystal fields on the expectation values may be "turned off" by superimposing a large magnetic field on the exchange field. We have done this explicitly for fields of 100 and 500 kOe. The limit as the external field becomes infinite (called hereafter the Zeeman limit or Z limit) is just the standard theory (see Sec. IV A) and is easily calculated analytically.

The reader may have noticed that our models are simply the four traceless terms which would be allowed in cubic symmetry. These should be regarded as 4 of the 24 (15+9) allowed orthorhombic terms for $\mathbf{F}^{\mathrm{ex}}$ and $\mathbf{G}$, we have chosen to investigate as a matter of taste. We emphasize that the crystal fields in the unstrained Hamiltonian were nine-parameter orthorhombic ones.

TABLE I. Levels of a representative Dy ion in crystal and exchange fields illustrating the extent of departure from Zeeman levels. The energy spacings are not uniform, the projection of J, and the squared projection, depart considerably from the m and m^2 ($m=\frac{15}{2}, \frac{13}{2}, \frac{11}{2}, \cdots$) rules. Nonetheless, the magnetostriction is often well given by the Z limit when plotted against $\langle J \rangle$.

Energy, cm^{-1}	$\langle J_z \rangle$	$\langle J_z^2 \rangle$
114.1	7.32	54.03
64.8	5.83	35.73
40.4	4.67	24.37
34.8	3.64	15.84
34.6	3.68	15.87
29.9	3.43	15.88
25.1	1.55	5.64
14.4	−0.05	1.52
−2.3	−0.92	3.54
−17.6	−2.91	15.15
−35.2	−4.43	31.15
−35.3	−2.18	9.85
−52.5	−3.80	19.85
−54.8	−3.90	17.90
−79.1	−5.85	34.84
−81.2	−6.17	38.83

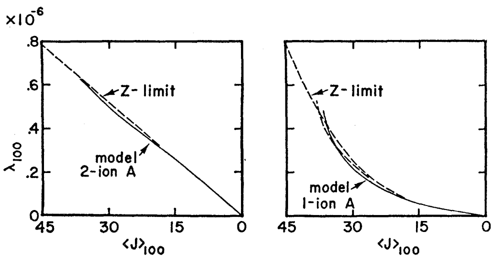

FIG. 3. λ_{100} versus $\langle J \rangle$ for A model. Solid line is for zero applied field, dashed line (labeled Z limit) is Zeeman (H— ∞) limit. The third line represents an intermediate case, H=100 kOe.

The extent to which the levels of Dy depart from Zeeman levels is illustrated in Table I. Here we give energies and quantum mechanical expectation values for J_z and $J_z{}^2$ for each of the 16 levels for a typical site. (Here the exchange field is along [001].) In the Z limit the levels are equally spaced and the expectation values go as m and m^2, respectively, where $m=-15/2$, $13/2$, $\cdots$. It is clear that one effect of the crystal field is to reduce the quantum-mechanical expectation values of both J_z and $J_z{}^2$ in the ground state and thus the size of $\mathcal{E}^{\mathrm{me}}$ and the magnetostriction at low temperature. The effect of temperature in reducing thermal expectation values is qualitatively the same and to the extent that this is so we may make the results for the actual case and the Z limit agree by plotting λ against the thermal expectation value of J along the field, $\langle J \rangle$, rather than against temperature. In the Z limit 2-ion λ's are linear in $\langle J \rangle$, and 1-ion λ's go as $\hat{I}_{5/2}$.

$\langle J \rangle (T)$ as computed from our molecular-field model summing over the six sites is given in Figs. 2(a) and 2(b) for the [100] and [111] directions, relevant to λ_{100} and λ_{111}, respectively. Note that even with averaging over sites and isotropic exchange, the crystal field makes the moment quite anisotropic at low tempera-

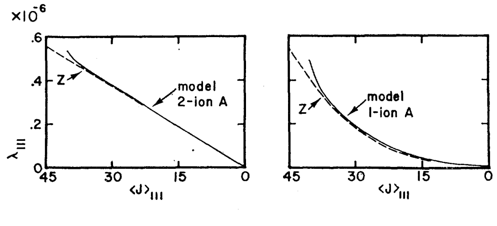

FIG. 4. λ_{111} versus $\langle J \rangle$ for A model, Z is Zeeman limit.

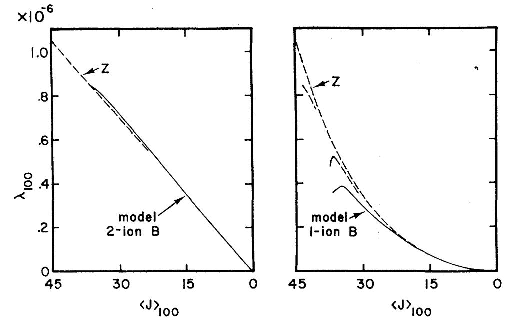

FIG. 5. λ_{100} versus $\langle J\rangle$ for B model. Solid line is $H=0$; dashes are $H=100$ kOe, $H=500$ kOe, and Z limit. The 1-ion model displays the most dramatic departure from the Z limit of the four models considered.

tures, [100] being 10% lower than [111] which in turn is about 10% below the Z limit of $45(=6\times15/2)$.

In Figs. 3–6 we give λ_{100} and λ_{111} for each of the four models. The values are plotted against $\langle J\rangle$ and thus the lines do not extend to the ordinate except in the Z limit. Note that near $\langle J\rangle=45$, the Z limit results reflect the isotropic theory being proportional to $\langle J\rangle$ for the 2-ion models and to $\langle J\rangle^3$ for 1-ion.

The effect of crystal fields on the temperature dependence of the λ's varies for each model. At higher temperatures (low $\langle J\rangle$) when many levels are contributing, crystal fields average out, but the Z limit is not approached particularly rapidly. The 1-ion B-model results for λ_{100} are interesting since here a fortuitous cancellation occurs at $T=0$°K which is lifted with rising temperature faster than the over-all decrease in the competing terms. Thus here is a case where λ actually rises as $\langle J\rangle$ falls (temperature increasing), a striking departure from the $\hat{I}_{5/2}$, Z limit theory.

There is a great deal of averaging over sites, levels, and expectation values implicit in this model and it is not surprising that by and large the crystal fields do not produce striking departures when plotted against $\langle J\rangle$. It is clear, however, that the low-temperature $\frac{1}{2}[l(l+1)]$ power law does not survive the introduction of substantial crystal fields.

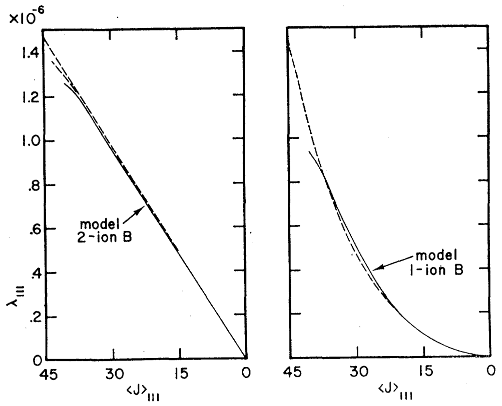

FIG. 6. λ_{111} versus $\langle J\rangle$ for B model. Unlike λ_{100}, here the 1-ion model is rather close to the Z limit.

The observed magnetostriction in DyIG[37] is peculiar in that λ_{111} follows an $\hat{I}_{5/2}$ (1-ion) temperature dependence rather nicely while λ_{100} is considerably below $\hat{I}_{5/2}$ at low temperature. The question then arises as to whether this is evidence that 2-ion terms are important for λ_{100}. The answer, from our model calculation, is no. The 1-ion B model has one λ follow $\hat{I}_{5/2}$ closely (Fig. 6) while the other λ does not (Fig. 4). Thus crystal field could explain the observed deviation from "usual" 1-ion behavior without recourse to 2-ion interactions.[44]

In conclusion, we have shown that when anisotropy, either in form of anisotropic exchange or crystal fields, is large, the relation of the underlying magnetoelastic spin Hamiltonian to magnetostriction becomes quite complex. Departure from the isotropic theory are to be expected; particularly at low temperatures.

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[44] Incidentally, the forced magnetostriction of DyIG (Ref. 37) shows no evidence of a contribution from a direct (F) term in fields up to 100 kOe.