

Sublattice Magnetization of Yttrium Iron Garnet at Low Temperatures*

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The energies and eigenvectors for long-wavelength acoustic spin waves in yttrium iron garnet are calculated neglecting spin-wave interactions, but including the effects of anisotropy and dipolar interactions. The sublattice magnetization is found to be $S_\alpha^z(T) = S - \delta_\alpha - cT - A_\alpha T^{3/2} - B_\alpha T^{5/2} - C_\alpha T^{7/2} \dots$. Here α labels the sublattice a or d , δ_α expresses the effect of zero-point motion, and cT is the Holstein-Primakoff correction for dipolar interactions. Expressions in terms of the exchange integrals J_{aa} , J_{dd} , J_{ad} , and J_{ad}' , where J_{ad}' describes interactions between next-nearest-neighbor a and d sites, are given for A_α , B_α , and, when $J_{ad}' = 0$, for C_α . The spin-wave spectrum of some substituted garnets and the effect of spin-wave interactions on the zero-point disordering are treated in appendices.

I. INTRODUCTION

THE use of nuclear magnetic resonance¹⁻⁴ as a tool for the investigation of magnetic materials has enabled one to make very accurate measurements of the temperature variation of the spontaneous magnetization of each magnetically inequivalent sublattice. The possibility of such measurements in turn stimulates detailed theoretical calculations of the sublattice magnetizations, especially for low temperatures where the spin-wave approximation is valid. Recently such accurate measurements have been performed on yttrium iron garnet (YIG)^{5,6}; so that by an accurate spin-wave analysis one may hope to deduce reliable values of the exchange coefficients. However, for YIG an earlier⁷ spin-wave analysis gave results in disagreement with both Wojtowicz's analysis⁸ of the high-temperature susceptibility data,⁹ and with evidence^{10,11} obtained from substituted garnets. This discrepancy has been largely removed by Gonano *et al.*¹² who have analyzed their magnetization data using the calculation we present here.

It is well known¹³ that, neglecting anisotropy and dipolar interactions, linearized spin-wave theory pre-

dicts the sublattice magnetization of a ferrimagnet to be of the form

$$M(T)/M(0) = 1 - \alpha T^{3/2} - \beta T^{5/2} - \gamma T^{7/2} \dots \quad (1.1)$$

The purpose of the present paper is to calculate the coefficients α , β , and γ for each sublattice in terms of the exchange coefficients. Previously⁷ we had obtained expressions for α and β in the analogous expansion for the total magnetization. In order to obtain the sublattice magnetization we study in more detail the transformation to normal modes. In addition we will calculate corrections to Eq. (1.1) when the effects of anisotropy, externally applied magnetic field, and dipolar interactions are included. We show that the simplest phenomenological extension of the Holstein-Primakoff¹⁴ results to the case of an anisotropic ferrimagnet is valid. We give these results since it has been shown^{5,12} that dipolar and anisotropy effects cannot be neglected when analyzing the low-temperature sublattice magnetizations of YIG.

In Appendix A we show that the ground-state value of the total magnetization of a ferrimagnet one obtains treating spin-wave interactions perturbatively is the same as the value in the Néel state. Appendix B is devoted to algebraic details involved in deriving expressions for the coefficients of Eq. (1.1). Finally, in Appendix C we give formulas for the spin-wave spectra of substituted garnets in which only the a or d sublattice is occupied by magnetic ions and antiferromagnetism results.

One may question the desirability of calculating analytically the terms in the expansion of Eq. (1.1) since the energies of all the normal modes (optical as well as acoustic) and hence the sublattice magnetizations could be calculated numerically on an electronic computer. Such a calculation is not trivial, however. In the first place the exchange integrals are not known precisely, so that the computer program would have to provide for evaluation of the sublattice magnetization over a range of parameters with a criterion for selecting the best fit to the experimental data.

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⁴A. J. Heeger and T. W. Houston, Phys. Rev. **135**, A661 (1964).

⁵R. Gonano, E. Hunt, H. Meyer, and A. B. Harris, J. Appl. Phys. **37**, 1322 (1966).

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⁷A. B. Harris, Phys. Rev. **132**, 2398 (1963).

⁸P. J. Wojtowicz, Phys. Letters **11**, 18 (1964).

⁹R. Aléonard, J. Phys. Chem. Solids **15**, 167 (1960).

¹⁰P. L. Richards and J. P. Remeika, J. Appl. Phys. **37**, 1310 (1966).

¹¹S. Geller, H. J. Williams, G. P. Espinosa, and R. C. Sherwood, Bell System Tech. J. **43**, 565 (1964).

¹²R. Gonano, E. Hunt, and H. Meyer (to be published).

¹³See, for instance, L. R. Walker in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, Chap. 8.

Also, for each set of parameters the eigenvalues of a 20×20 matrix must be evaluated for a mesh of points in reciprocal space. Such a computation would be time-consuming and would involve sophisticated programming. In contrast, our results were obtained with only a few weeks labor and in fact appear to be sufficient, at least at low temperatures, for the analysis of the experimental data, as is explained by Gonano *et al.*¹²

II. CALCULATION OF THE SPIN-WAVE SPECTRUM OF YTTRIUM IRON GARNET

The notation we use is as follows. R is a vector of the bcc lattice

$$\mathbf{R} \equiv \mathbf{R}(n_1, n_2, n_3) = \frac{1}{2}a(n_1\hat{i} + n_2\hat{j} + n_3\hat{k}), \quad (2.1)$$

where a is the lattice constant, \hat{i} , \hat{j} , and \hat{k} are unit vectors in the x , y , and z directions, respectively, and n_1 , n_2 , and n_3 are integers either all even or all odd. Let $\mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(na)}$ denote the spin operator associated with the lattice site at $\mathbf{R}+\boldsymbol{\tau}(na)$, where $\boldsymbol{\tau}(na)$ is the position of the n th a site ($n=1, 8$) within the unit cell. Let $\mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(nd)}$ denote the spin operator associated with the lattice site at $\mathbf{R}+\boldsymbol{\tau}(nd)$, where $\boldsymbol{\tau}(nd)$ is the position of the n th d site ($n=1, 12$) within the unit cell. The labeling of the a and d sites in the unit cell is the same as in a previous treatment.⁷

Initially we treat the case of isotropic exchange interactions governed by a Heisenberg Hamiltonian. The application of linearized spin-wave theory to such a system is well known.¹³ We give here a brief review mainly to fix the notation. For YIG we take the Hamiltonian to be

$$\begin{aligned} \mathcal{H} = & -J_{aa} \sum_{nn} \mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(na)} \cdot \mathbf{S}_{\mathbf{R}'+\boldsymbol{\tau}'(n'a)} \\ & -J_{dd} \sum_{nn} \mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(nd)} \cdot \mathbf{S}_{\mathbf{R}'+\boldsymbol{\tau}'(n'd)} \\ & -2J_{ad} \sum_{nn} \mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(na)} \cdot \mathbf{S}_{\mathbf{R}'+\boldsymbol{\tau}'(n'd)} \\ & -2J_{ad'} \sum_{nn} \mathbf{S}_{\mathbf{R}+\boldsymbol{\tau}(na)} \cdot \mathbf{S}_{\mathbf{R}'+\boldsymbol{\tau}'(n'd)}, \end{aligned} \quad (2.2)$$

where the subscripts nn indicate that the first three summations are taken over \mathbf{R} , \mathbf{R}' , n , and n' such that $\mathbf{R}+\boldsymbol{\tau}$ and $\mathbf{R}'+\boldsymbol{\tau}'$ are nearest-neighbor pairs of lattice sites in the sublattice in question. The last term of Eq. (2.2) is summed over values of \mathbf{R} , \mathbf{R}' , n , and n' such that $\mathbf{R}+\boldsymbol{\tau}$ and $\mathbf{R}'+\boldsymbol{\tau}'$ are next-nearest-neighbor pairs of ions in the a and d sublattices. The exchange interactions are governed by the coupling constants J_{aa} , J_{ad} , J_{dd} , and $J_{ad'}$. The effect of dipolar interactions and crystalline anisotropy will be discussed in Sec. III. We remark that the Hamiltonian (2.2) is invariant under rotations so that the various a (or d) sites which are completely equivalent to each other with respect to nonmagnetic interactions will remain equivalent when

the Hamiltonian (2.2) is considered. Therefore the magnetization is expected to be the same throughout the entire a (or d) sublattice. When dipolar interactions are considered, this argument breaks down because the Hamiltonian is not invariant under rotation of the direction of magnetization.^{15,16} However, this effect is probably too small to be observed.

The simplest spin-wave theory which neglects spin-wave interactions can be obtained by substituting boson operators for the spin operators at each site as follows:

$$S^z = \mp [S - a^\dagger a], \quad (2.3a)$$

$$S^\pm = (2S)^{1/2} a^\dagger, \quad (2.3b)$$

$$S^\mp = (2S)^{1/2} a, \quad (2.3c)$$

where we take the upper choice of sign for the a sites and the lower choice of sign for the d sites. We transform to momentum variables

$$a_{na}^\dagger(\mathbf{k}) = N_{uc}^{-1/2} \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot [\mathbf{R} + \boldsymbol{\tau}(na)]) a_{na}^\dagger(\mathbf{R}), \quad (2.4a)$$

$$a_{nd}^\dagger(\mathbf{k}) = N_{uc}^{-1/2} \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot [\mathbf{R} + \boldsymbol{\tau}(nd)]) a_{nd}^\dagger(\mathbf{R}), \quad (2.4b)$$

where N_{uc} is the number of bcc unit cells, so that

$$N_{uc} = \sum_{\mathbf{k}} 1; \quad V = \frac{1}{2} N_{uc} a^3. \quad (2.5)$$

Using the substitutions (2.3) and (2.4) in the Hamiltonian of Eq. (2.2) we find

$$\begin{aligned} \mathcal{H} = & \sum_{\mathbf{k}, n, n'} M_{nn'}^{aa}(\mathbf{k}) a_{an}^\dagger(\mathbf{k}) a_{an'}(\mathbf{k}) \\ & + \sum_{\mathbf{k}, n, n'} M_{nn'}^{dd}(\mathbf{k}) a_{dn}^\dagger(\mathbf{k}) a_{dn'}(\mathbf{k}) \\ & + \sum_{\mathbf{k}, n, n'} \{ M_{nn'}^{ad}(\mathbf{k}) a_{an}^\dagger(\mathbf{k}) a_{dn'}^\dagger(-\mathbf{k}) \\ & + [M_{nn'}^{ad}(\mathbf{k})]^* a_{an}(\mathbf{k}) a_{dn'}(-\mathbf{k}) \}, \end{aligned} \quad (2.6)$$

TABLE I. The matrix $\mathbf{M}^{aa}(\mathbf{k})$. Here $A = 40J_{aa} - 30J_{ad} - 30J'_{ad}$ and $(lmn) = -10J_{aa} \cos([\mathbf{k}_x + m\mathbf{k}_y + n\mathbf{k}_z]a/8)$.

$n' \backslash n$	1	2	3	4	5	6	7	8
1	A	0	0	0	(222)	($\bar{2}\bar{2}\bar{2}$)	($2\bar{2}\bar{2}$)	($\bar{2}2\bar{2}$)
2	0	A	0	0	($\bar{2}\bar{2}\bar{2}$)	(222)	(222)	($\bar{2}\bar{2}\bar{2}$)
3	0	0	A	0	(222)	($\bar{2}\bar{2}\bar{2}$)	(222)	($\bar{2}\bar{2}\bar{2}$)
4	0	0	0	A	($\bar{2}\bar{2}\bar{2}$)	(222)	($\bar{2}\bar{2}\bar{2}$)	(222)
5	(222)	($\bar{2}\bar{2}\bar{2}$)	($\bar{2}\bar{2}\bar{2}$)	($\bar{2}\bar{2}\bar{2}$)	A	0	0	0
6	($\bar{2}\bar{2}\bar{2}$)	(222)	(222)	(222)	0	A	0	0
7	($\bar{2}\bar{2}\bar{2}$)	(222)	($\bar{2}\bar{2}\bar{2}$)	($\bar{2}\bar{2}\bar{2}$)	0	0	A	0
8	(222)	($\bar{2}\bar{2}\bar{2}$)	(222)	(222)	0	0	0	A

¹⁵ J. O. Dimmock and R. G. Wheeler, Phys. Rev. **127**, 39 (1962).

¹⁶ W. Brinkman and R. J. Elliott, J. Appl. Phys. **37**, 1457 (1966).

Eq. (2.9) we obtain

$$\begin{pmatrix} \mathbf{M}^{aa}(\mathbf{k}) & -\mathbf{M}^{ad}(\mathbf{k}) \\ [\mathbf{M}^{ad}(\mathbf{k})]^\dagger & -\tilde{\mathbf{M}}^{dd}(-\mathbf{k}) \end{pmatrix} \begin{pmatrix} \mathbf{I}_\rho^{aa}(\mathbf{k}) \\ \mathbf{I}_\rho^{da}(\mathbf{k}) \end{pmatrix} \\ = \hbar\omega_\rho(\mathbf{k}) \begin{pmatrix} \mathbf{I}_\rho^{aa}(\mathbf{k}) \\ \mathbf{I}_\rho^{da}(\mathbf{k}) \end{pmatrix}, \quad (2.11a)$$

$$\begin{pmatrix} \mathbf{M}^{aa}(\mathbf{k}) & -\mathbf{M}^{ad}(\mathbf{k}) \\ [\mathbf{M}^{ad}(\mathbf{k})]^\dagger & -\tilde{\mathbf{M}}^{dd}(-\mathbf{k}) \end{pmatrix} \begin{pmatrix} \mathbf{I}_\rho^{ad}(\mathbf{k}) \\ \mathbf{I}_\rho^{dd}(\mathbf{k}) \end{pmatrix} \\ = -\hbar\omega_\rho(-\mathbf{k}) \begin{pmatrix} \mathbf{I}_\rho^{ad}(\mathbf{k}) \\ \mathbf{I}_\rho^{dd}(\mathbf{k}) \end{pmatrix}. \quad (2.11b)$$

Here \sim means transpose, \dagger means Hermitian conjugate, and \mathbf{I}_ρ^{aa} , etc. are column vectors. In solving (2.11), if $\hbar\omega_\rho(\mathbf{k})$ is negative then one has found the transformation to $\mathbf{Q}_\rho(-\mathbf{k})$ since (2.11b) differs from (2.11a) only in the sign of $\hbar\omega_\rho(\mathbf{k})$. Hence the columns of

$$\mathbf{L} \equiv \begin{pmatrix} \mathbf{I}^{aa} & \mathbf{I}^{ad} \\ \mathbf{I}^{da} & \mathbf{I}^{dd} \end{pmatrix} \quad (2.12)$$

are the right eigenvectors of the matrix in (2.11) and give the transformation from the $a_{an}(\mathbf{k})$ and $a_{dn}(-\mathbf{k})$ to the normal modes. The left eigenvectors are the rows of

$$\mathbf{L}^t \equiv \begin{pmatrix} [\mathbf{I}^{aa}]^\dagger & -[\mathbf{I}^{da}]^\dagger \\ -[\mathbf{I}^{ad}]^\dagger & [\mathbf{I}^{dd}]^\dagger \end{pmatrix}, \quad (2.13)$$

since $\mathbf{M}^{aa}(\mathbf{k})$ and $\mathbf{M}^{dd}(\mathbf{k})$ are Hermitian. The normalization of these eigenvectors is such that $\mathbf{L}^t\mathbf{L}=\mathbf{1}$, since we demand $[\mathbf{Q}_\rho(\mathbf{k}), \mathbf{Q}_{\rho'}(\mathbf{k}')^\dagger] = \delta_{\rho\rho'}\delta_{\mathbf{k}\mathbf{k}'}$. The matrix \mathbf{L}^t therefore provides the inverse transformation which is what we wish to find. Using these row vectors one can express the $a_{an}(\mathbf{k})$ and $a_{dn}(\mathbf{k})$ which create spin deviations on a particular sublattice in terms of the normal mode operators.

However, since the magnetization of all the sublattices of each type (a or d) is the same, we can obtain a simplification by computing the total magnetization of all the a and d sublattices, respectively. To do this it is convenient to add to the Hamiltonian a term V containing the indicator fields H_a and H_d :

$$V = -g\beta \sum_{\mathbf{R}} [H_a \sum_{n=1,8} S_{\mathbf{R}+\tau(na)}^\varepsilon + H_d \sum_{n=1,12} S_{\mathbf{R}+\tau(nd)}^\varepsilon]. \quad (2.14)$$

Then one finds the total sublattice magnetizations at temperature T as

$$\langle M_a \rangle_T = \frac{\partial F}{\partial H_a} \Big|_T; \quad \langle M_d \rangle_T = -\frac{\partial F}{\partial H_d} \Big|_T, \quad (2.15)$$

where F is the free energy, and the derivatives are

evaluated for $H_a=H_d=0$. Explicitly, Eq. (2.15) is

$$\frac{\langle S_a \rangle_T}{S} \equiv \frac{\langle M_a \rangle_T}{8N_{uc}g\beta S} = 1 - \delta_a - (8N_{uc}g\beta S)^{-1} \sum_{\mathbf{k},\rho} \left| \frac{\partial \hbar\omega_\rho(\mathbf{k})}{\partial H_a} \right| \\ \times \{ \exp(\hbar\omega_\rho(\mathbf{k})/k_B T) - 1 \}^{-1}, \quad (2.16a)$$

$$\frac{\langle S_d \rangle_T}{S} \equiv \frac{\langle M_d \rangle_T}{12N_{uc}g\beta S} = 1 - \delta_d - (12N_{uc}g\beta S)^{-1} \sum_{\mathbf{k},\rho} \left| \frac{\partial \hbar\omega_\rho(\mathbf{k})}{\partial H_d} \right| \\ \times \{ \exp(\hbar\omega_\rho(\mathbf{k})/k_B T) - 1 \}^{-1}. \quad (2.16b)$$

Note that excitation of any mode decreases the sublattice magnetizations, whereas the total magnetization decreases or increases according to the choice of sign in

$$\left| \frac{\partial \hbar\omega_\rho(\mathbf{k})}{\partial H_a} \right| - \left| \frac{\partial \hbar\omega_\rho(\mathbf{k})}{\partial H_d} \right| = \pm g\beta. \quad (2.17)$$

Because of zero-point motion $\langle S_a \rangle_T$ and $\langle S_d \rangle_T$ are somewhat less than their values in the Néel state. Walker,¹³ however, has pointed out that the total magnetization at zero temperature is given by its value in the Néel state, which implies that

$$\delta_a = \frac{2}{3}\delta_d \equiv \delta. \quad (2.18)$$

As we discuss in Appendix A, spin-wave interactions do not affect this relationship.

Recently, Brinkman and Elliott¹⁶ have pointed out that the spin-wave matrix equation, (2.11), becomes simpler when \mathbf{k} lies along certain symmetry directions. In a previous treatment⁷ we overlooked some of the symmetry operations and therefore did not obtain the maximum simplification. As a result our numerical computations were more time consuming than necessary. In order to determine analytic expressions for the coefficients in Eqs. (2.21) and (2.22) below, one needs to calculate the acoustic eigenvalue for at least three orientations of \mathbf{k} . Rather than make separate calculations for each direction of \mathbf{k} , we will undertake one calculation for general orientation of wave vector using perturbation theory. In order to obtain analytic results, we will henceforth completely neglect the optical modes. Thus our results will be valid at temperatures such that $k_B T$ is much less than the energy of the lowest optical mode. By replacing the optical modes by a suitable number of wave-vector-independent (Einstein) oscillators, one may profitably use our results at somewhat higher temperatures.

Our procedure is as follows: We denote the matrix in Eq. (2.11) by $\mathfrak{M}(\mathbf{k})$ and expand it in powers of the components k^μ ($\mu=x, y, \text{ or } z$) as

$$\mathfrak{M}(\mathbf{k}) = \sum_n \mathfrak{M}_n, \quad (2.19)$$

where \mathfrak{M}_n varies as $|\mathbf{k}|^n$:

$$\mathfrak{M}_0 = \mathfrak{M}(0); \quad \mathfrak{M}_1 = \sum_\mu k^\mu \frac{\partial}{\partial k^\mu} \mathfrak{M}(\mathbf{k}) \Big|_{\mathbf{k}=0}; \text{ etc.} \quad (2.20)$$

We take \mathfrak{M}_0 , whose eigenvectors and eigenvalues are known,¹⁷ to be the unperturbed Hamiltonian, and treat the remaining terms in Eq. (2.19) as the perturbation. The formula¹⁸ for an eigenvalue inclusive of fourth-order terms in the perturbation can be used with the slight modification that since $\mathfrak{M}(\mathbf{k})$ is not Hermitian, bras and kets in the formula should be interpreted as left and right eigenvectors according to the definitions of Eqs. (2.12) and (2.13), respectively. Because of the cubic symmetry¹⁹ one thus obtains the acoustic-mode energy in the form

$$\begin{aligned} \hbar\omega_{ac} = & DK^2 + EK^4 + F(K_x^2K_y^2 + K_x^2K_z^2 + K_y^2K_z^2) \\ & + G(K_x^6 + K_y^6 + K_z^6) + HK^2(K_x^4 + K_y^4 + K_z^4) \\ & + IK_x^2K_y^2K_z^2 \dots, \end{aligned} \quad (2.21)$$

where $\mathbf{K} \equiv \mathbf{ak}$. Likewise one obtains the expansion

$$\begin{aligned} \frac{\partial \hbar\omega_{ac}}{\partial H_a} = & -2g\beta\{1 - AK^2 + \beta K^4 \\ & + C(K_x^2K_y^2 + K_x^2K_z^2 + K_y^2K_z^2) \dots\}, \end{aligned} \quad (2.22)$$

which can be combined with Eq. (2.17) to find $\partial \hbar\omega_{ac}/\partial H_a$. Substituting these expansions into Eq.

(2.16) we find the sublattice magnetization to be

$$\begin{aligned} \frac{\Delta S_\nu}{S_\nu} = \frac{S - \langle S_\nu \rangle_T}{S} = & \delta_\nu + \frac{2}{5}\zeta\left(\frac{3}{2}\right) \left(\frac{k_B T}{D}\right)^{3/2} \\ & \times \left[1 + \beta_\nu' \left(\frac{k_B T}{D}\right) + \gamma_\nu' \left(\frac{k_B T}{D}\right)^2 + \dots \right], \end{aligned} \quad (2.23)$$

where

$$\beta_\nu' = -\frac{\zeta\left(\frac{5}{2}\right) \times 1}{\zeta\left(\frac{3}{2}\right) \times 4D} \{15E + 3F + 6\xi_\nu AD\}, \quad (2.24a)$$

$$\begin{aligned} \gamma_\nu' = \frac{\zeta\left(\frac{7}{2}\right)}{\zeta\left(\frac{5}{2}\right)} \frac{1}{32D^2} \{ & 945E^2 + 378EF + 45F^2 - 180DG \\ & - 252DH - 4DI + 420\xi_\nu AED + 84\xi_\nu AFD \\ & + 120\xi_\nu BD^2 + 24\xi_\nu CD^2 \}, \end{aligned} \quad (2.24b)$$

where $\zeta(n)$ is the Riemann zeta function, ν assumes the values a and d , and

$$\xi_a = 1, \quad \xi_d = \frac{2}{3}. \quad (2.25)$$

It remains to calculate the coefficients A, B, C , etc.¹⁹ introduced in Eqs. (2.12) and (2.22). The acoustic mode energy is given by the perturbation series¹⁸

$$\begin{aligned} -\hbar\omega_{ac}(\mathbf{k}) = & \langle 0 | \mathfrak{M}_2 | 0 \rangle + \langle 0 | \mathfrak{M}_4 | 0 \rangle - \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n} + \langle 0 | \mathfrak{M}_6 | 0 \rangle - 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathfrak{M}_4 | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n} \\ & - \sum' \frac{\langle 0 | \mathfrak{M}_3 | n \rangle \langle n | \mathfrak{M}_3 | 0 \rangle}{E_n} + \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_2 | m \rangle \langle m | \mathfrak{M}_2 | 0 \rangle}{E_n E_m} + 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_1 | m \rangle \langle m | \mathfrak{M}_3 | 0 \rangle}{E_n E_m} \\ & - \sum' \frac{\langle 0 | \mathfrak{M}_2 | 0 \rangle \langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n^2} - \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_1 | l \rangle \langle l | \mathfrak{M}_1 | m \rangle \langle m | \mathfrak{M}_2 | 0 \rangle}{E_n E_m E_l}, \end{aligned} \quad (2.26)$$

where primes indicate omission of terms with zero denominators, $|0\rangle$ is the acoustic eigenvector for $k=0$ which has zero energy, and $|n\rangle$ an optical mode eigenvector for $k=0$ with eigenvalue E_n . We have already simplified this formula by taking account of the cubic symmetry and the fact that

$$\mathfrak{M}_1 | 0 \rangle = 0. \quad (2.27)$$

Similarly, we find

$$\begin{aligned} \frac{1}{g\beta} \frac{\partial \hbar\omega_{ac}(k)}{\partial H_d} = & \langle 0 | \mathbf{1}_d | 0 \rangle - 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathbf{1}_d | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n} - 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathbf{1}_d | n \rangle \langle n | \mathfrak{M}_4 | 0 \rangle}{E_n} \\ & + 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathbf{1}_d | n \rangle \langle n | \mathfrak{M}_2 | m \rangle \langle m | \mathfrak{M}_2 | 0 \rangle}{E_n E_m} + \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathbf{1}_d | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n^2} \\ & - \sum' \frac{\langle 0 | \mathfrak{M}_2 | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle \langle 0 | \mathbf{1}_d | 0 \rangle}{E_n^2} - 2 \operatorname{Re} \sum' \frac{\langle 0 | \mathfrak{M}_2 | 0 \rangle \langle 0 | \mathbf{1}_d | n \rangle \langle n | \mathfrak{M}_2 | 0 \rangle}{E_n^2}, \end{aligned} \quad (2.28)$$

where $\mathbf{1}_d$ is a unit operator in the space of d sublattice excitations. In Appendix B we tabulate the eigenvectors,

¹⁷ R. L. Douglass, Phys. Rev. **120**, 1612 (1960); B. Dreyfus, J. Phys. Chem. Solids **23**, 287 (1962).

¹⁸ K. F. Niessen, Phys. Rev. **34**, 253 (1929).

¹⁹ We here give expressions only for β_ν' and γ_ν' . The expression for A is given in Ref. 5 and those for E and F when $J_{ad}'=0$ are given in Ref. 7. We point out an inconsistency in the signs of Eqs. (16), (17), and (18) in Ref. 7; namely, the left-hand side of these equations should be $-D$, $-E$, and $-F$, respectively.

eigenvalues, and matrix elements required for the evaluation of Eqs. (2.26) and (2.28). Thus we find

$$\beta_{\nu'} = \left[\zeta\left(\frac{5}{2}\right)/4\zeta\left(\frac{3}{2}\right)D \right] \left\{ \frac{3}{2}\xi_{\nu}D \left[\frac{-48J_{aa} + 25J_{ad} + 65J_{ad'} - 12J_{dd}}{16(J_{ad} + J_{ad'})} \right] + \frac{5}{4096} \left[-288J_{aa} + 75J_{ad} - 54J_{dd} + 507J_{ad'} \right] \right. \\ \left. + 5(-48J_{aa} + 25J_{ad} + 65J_{ad'} - 12J_{dd})^2/(J_{ad} + J_{ad'}) + 2(3J_{dd} - 2J_{ad} + 14J_{ad'})^2/(2J_{ad} + 2J_{ad'} - 3J_{dd}) \right. \\ \left. + 48(3J_{ad'} - J_{ad})^2/(3J_{ad} + 3J_{ad'} - 4J_{aa}) \right\}, \quad (2.29)$$

and for $J_{ad'}=0$,

$$32\gamma_{\nu'}\zeta\left(\frac{3}{2}\right)/\zeta\left(\frac{5}{2}\right) = 105 \left\{ -\frac{1}{256} \frac{(25-48x-12y)^2}{(5-8x-3y)} + \frac{1}{256} \frac{1}{(5-8x-3y)} - \frac{1}{64} \right\} \left\{ -\frac{1}{256} \frac{(25-48x-12y)^2}{(5-8x-3y)} + \frac{1}{256} \frac{1}{(5-8x-3y)} \right. \\ \left. - \frac{1}{64} \frac{(5x-8x^2)}{(6-8x)(25-40x-15y)} \right\} + \frac{5}{16} \left[\frac{5x-8x^2}{(3-4x)(5-8x-3y)} \right]^2 - \frac{21}{128(3-4x)^2} - \frac{175}{2048} \frac{(25-48x-12y)^2(2-4x-y)}{(5-8x-3y)} \\ - \frac{175}{8192} (25-48x-12y) - \frac{7(25-48x-12y)}{192(5-8x-3y)} \left[\frac{19}{32} - \frac{25}{3}x \right] + \frac{3}{2}\xi_{\nu} \left\{ \frac{(125-576x-72y)}{256} + \frac{1}{16(3-4x)} + \frac{1-y}{128} + \frac{3}{16(3-4x)^2} \right. \\ \left. - \frac{1}{768} + \frac{100}{3} \left(\frac{25-48x-12y}{64} \right)^2 + \frac{25}{128} (2-4x-y)(25-48x-12y) \right\}, \quad (2.30)$$

where

$$D = (40J_{aa} - 25J_{ad} - 65J_{ad'} + 15J_{dd})/16, \quad (2.31a)$$

$$x = J_{aa}/J_{ad}, \quad (2.31b)$$

$$y = J_{dd}/J_{ad}. \quad (2.31c)$$

We remark that our results agree with, where they overlap, previous numerical and analytical results.⁷ In Table IV we have summarized these results by giving numerical values of $\beta_{\nu'}$ and $\gamma_{\nu'}$ for the cases $J_{ad'}=0$ as a function of x and y . Using these numerical results curves of the sublattice and total magnetizations are readily constructed. An experimental value for D can be very accurately determined in this way providing dipolar interactions and anisotropy are not neglected.^{5,12} From Table IV we see that unfortunately the low-temperature magnetization curves are not very sensitive to variations in y , so that from experimental values

of the coefficients $\beta_{\nu'}$ and $\gamma_{\nu'}$ one can probably only determine x . However, when the contribution of the optical modes is taken into account the magnetization curves will depend on y , thus enabling its determination.

Finally we comment that our previous⁷ erroneous conclusion from analysis of low-temperature magnetization data²⁰ that $J_{aa}/J_{ad} \approx 0.2$ was due to a poor choice for the parameter D . Had we chosen the value recently deduced from the high-resolution nuclear magnetic resonance data,¹² we would have obtained a much smaller value of J_{aa}/J_{ad} in agreement with studies^{10,11} of substituted garnets. It is perhaps relevant to point out that a high-temperature analysis such as that performed by Wojtowicz⁸ is expected to give somewhat different values of the exchange integrals than those obtained from a spin-wave analysis due to thermal expansion. According to Bloch *et al.*²¹ exchange integrals in the iron garnets vary as the $-(10/3)$

TABLE IV. Values of $\beta_{\nu'}$ and $\gamma_{\nu'}$ for $J_{ad'}=0$. $x=J_{aa}/J_{ad}$ and $y=J_{dd}/J_{ad}$.

$10\beta_{a'}$					$10\beta'_{a'}$				
0.3			0.026		0.3			0.501	
0.2	0.287	0.144	0.003	-0.122	0.2	1.20	0.858	0.526	0.204
0.1	0.255	0.113	-0.020		0.1	1.21	0.877	0.549	
0.0	0.222	0.087	-0.043		0.0	1.22	0.897	0.575	
y/x	0.0	0.1	0.2	0.3	y/x	0.0	0.1	0.2	0.3
$10^3\gamma_{a'}$					$10^3\gamma'_{a'}$				
0.3			+0.2		0.3			4.4	
0.2	-3.0	-1.7	-0.2	+1.3	0.2	18.2	9.7	4.2	1.5
0.1	-4.6	-2.5	-0.6		0.1	17.5	9.3	4.2	
0.0	-6.1	-3.6	-0.9		0.0	16.3	8.5	3.8	
y/x	0.0	0.1	0.2	0.3	y/x	0.0	0.1	0.2	0.3

²⁰ I. H. Solt, Jr., J. Appl. Phys. 33, 1189S (1962).

²¹ D. Bloch, F. Chaissé, and R. Pauthenet, J. Appl. Phys. 37, 1401 (1966).

power of the volume. Using the observed²² temperature variation of the lattice constant, we have thus estimated that the exchange integrals in YIG are approximately 10% less at 800°K than at zero temperature. Accordingly, from Wojtowicz's analysis one would predict $J_{ad} = -24.6 \text{ cm}^{-1}$, $J_{dd} = -2.5 \text{ cm}^{-1}$, and $J_{aa} = -0.2 \text{ cm}^{-1}$ at zero temperature. These values give a value of D some 15% larger than that obtained by the high-resolution nuclear-magnetic-resonance data. This remaining discrepancy may be caused by inaccuracies in Aléonard's⁹ susceptibility data due to impurities. As evidence for this statement we cite the discrepancy between Anderson's²³ susceptibility measurements and those of Aléonard. Also, as Aléonard has indicated, the volume dependence of the exchange integrals implies a temperature dependence of the form $J = J_0(1 - \gamma T)$ where $\gamma \sim 10^{-4}$ per °K. Thus thermal expansion has the effect of renormalizing the coefficients given by Wojtowicz for temperature-independent exchange integrals. Accordingly it is quite possible that use of more reliable high-temperature susceptibility data in a more refined analysis would resolve the discrepancy between the interpretations of high-temperature and low-temperature data.

III. THE EFFECT OF DIPOLAR INTERACTIONS, ANISOTROPY, AND EXTERNAL MAGNETIC FIELD ON THE SUBLATTICE MAGNETIZATION

In this section we discuss the modifications necessary to take account of dipolar interactions, crystalline anisotropy, and externally applied magnetic field.

We first discuss the effect of dipolar interactions. The Hamiltonian for dipolar interactions is

$$\mathcal{H}_D = \frac{1}{2} \sum_{ij} \left\{ \frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - \frac{3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \right\} g_i g_j \beta^2 \quad (3.1)$$

in the usual notation. If we use the substitutions (2.3) and (2.4) in this Hamiltonian and neglect terms higher than quadratic in the boson operators, we obtain an effective dipolar Hamiltonian of the form

$$\begin{aligned} \mathcal{H}_{\text{eff}} = \sum_{\mathbf{k}} \sum_{nn'} \{ & C_{nn'}(\mathbf{k}) a_n^\dagger(\mathbf{k}) a_{n'}(\mathbf{k}) \\ & + D_{nn'}(\mathbf{k}) a_n^\dagger(\mathbf{k}) a_{n'}^\dagger(-\mathbf{k}) \\ & + [D_{nn'}(\mathbf{k})]^* a_n(\mathbf{k}) a_{n'}(-\mathbf{k}) \}, \quad (3.2) \end{aligned}$$

where n and n' are summed over all the a and d sublattices. The omission of terms higher than quadratic in the boson operators corresponds to the neglect of spin-wave interactions and is consistent with the accuracy of the calculations of the previous section. The constant term and terms linear in the boson

operators have also been omitted in Eq. (3.2), since they do not effect the frequencies of the normal modes. We assume that the dipolar wave sums, $C_{nn'}(\mathbf{k})$ etc., which appear in Eq. (3.2) are translationally invariant. This approximation is expected to be valid²⁴ unless $1/k$ is comparable to the sample dimensions. The lattice sums, $C_{nn'}(\mathbf{k})$ etc., are in general complicated and are not readily evaluated. However, an important simplification occurs if we attempt a solution of the normal mode problem for small k and we keep only those terms which are smaller than the exchange terms by order (M/H_E) where H_E is a typical exchange field. Such an approximation is obtained by substituting for the $a_n^\dagger(\mathbf{k})$ the expressions for $k=0$ for the transformation to the normal modes neglecting dipolar interactions. We can neglect the k dependence of the transformation coefficients because terms of order k^2 lead to corrections which are smaller than those we consider by a factor on the order of $(k_B T / g\beta H_E)$. Thus \mathcal{H}_{eff} takes the form

$$\begin{aligned} \mathcal{H}_{\text{eff}} = \sum_{\mathbf{k}} \sum_{\rho\rho'} \{ & \bar{C}_{\rho\rho'}(\mathbf{k}) \mathbf{Q}_\rho^\dagger(\mathbf{k}) \mathbf{Q}_\rho(\mathbf{k}) \\ & + \bar{D}_{\rho\rho'}(\mathbf{k}) \mathbf{Q}_\rho^\dagger(\mathbf{k}) \mathbf{Q}_{\rho'}^\dagger(-\mathbf{k}) \\ & + [\bar{D}_{\rho\rho'}(\mathbf{k})]^* \mathbf{Q}_\rho(\mathbf{k}) \mathbf{Q}_{\rho'}(-\mathbf{k}) \}. \quad (3.3) \end{aligned}$$

As is well known, the first-order energy shifts are found by keeping only the semidiagonal part of \mathbf{H}_{eff} . Thus for the optical modes the effect of dipolar interactions^{15,16} is to reduce the degeneracy of the optical modes. For the acoustic mode one thus has the following effective Hamiltonian:

$$\begin{aligned} \mathcal{H}_{\text{eff}} \sim \sum_{\mathbf{k}} \{ & C_0(\mathbf{k}) \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0(\mathbf{k}) + D_0(\mathbf{k}) \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0^\dagger(-\mathbf{k}) \\ & + [D_0(\mathbf{k})]^* \mathbf{Q}_0(\mathbf{k}) \mathbf{Q}_0(-\mathbf{k}) \}, \quad (3.4) \end{aligned}$$

where

$$\begin{aligned} C_0(\mathbf{k}) = \frac{1}{8} g^2 \beta^2 S \sum_{\tau, \tau', \mathbf{R}'} \frac{(3 \cos^2 \theta - 1) S_{\tau^z}}{|\mathbf{R}' + \boldsymbol{\tau}' + \boldsymbol{\tau}|^3 S_{\tau'^z}} \\ \times \{ 2 + \exp(-i\mathbf{k} \cdot [\boldsymbol{\tau} - \mathbf{R}' - \boldsymbol{\tau}']) \}, \quad (3.5a) \end{aligned}$$

$$\begin{aligned} D_0(\mathbf{k}) = -\frac{3}{16} g^2 \beta^2 S \sum_{\tau, \tau', \mathbf{R}'} \frac{\sin^2 \theta \exp(2i\varphi) S_{\tau^z}}{|\mathbf{R}' + \boldsymbol{\tau}' - \boldsymbol{\tau}|^3 S_{\tau'^z}} \\ \times \{ \exp(-i\mathbf{k} \cdot [\boldsymbol{\tau} - \mathbf{R}' - \boldsymbol{\tau}']) \}. \quad (3.5b) \end{aligned}$$

In Eq. (3.5) the sum is over all sites $\boldsymbol{\tau}$ and $\boldsymbol{\tau}'$ within the unit cell and over all \mathbf{R}' except that $\mathbf{R}' + \boldsymbol{\tau}' = \boldsymbol{\tau}$ is excluded. S_{τ^z} is the value of the z component of spin of the lattice site at $\boldsymbol{\tau}$ in the Néel state ($S_{\tau^z} = \pm \frac{5}{2}$). Also

$$\cos \theta = (\mathbf{R}' + \boldsymbol{\tau}' - \boldsymbol{\tau}) \cdot \hat{z} / |\mathbf{R}' + \boldsymbol{\tau}' - \boldsymbol{\tau}|, \quad (3.6a)$$

$$\sin \theta \cos \varphi = (\mathbf{R}' + \boldsymbol{\tau}' - \boldsymbol{\tau}) \cdot \hat{x} / |\mathbf{R}' + \boldsymbol{\tau}' - \boldsymbol{\tau}|, \quad (3.6b)$$

where \hat{x} and \hat{z} are unit vectors along the x and z axes, respectively. Evaluating these lattice sums in the

²² S. Geller and M. A. Gilleo, J. Phys. Chem. Solids 3, 30 (1957).

²³ E. E. Anderson, Phys. Rev. 134, A1581 (1964).

²⁴ M. H. Cohen and F. Keffer, Phys. Rev. 99, 1128 (1955).

manner of Holstein and Primakoff,¹⁴ we find

$$C_0(\mathbf{k}) = -g\beta N_z M + 2\pi g\beta M \sin^2 \theta_k, \quad (3.7a)$$

$$D_0(\mathbf{k}) = \pi g\beta M \sin^2 \theta_k, \quad (3.7b)$$

where $\cos \theta_k = \mathbf{k} \cdot \hat{z} / |\mathbf{k}|$ and M is the magnetic moment per unit volume. Thus we obtain results identical to those for a single sublattice. This does not surprise us, since for long wavelength all the sublattices are in phase and the spin-wave energy is independent of structure assuming cubic symmetry. As we have argued previously,²⁵ when there is no external field the domain arrangement is such that the demagnetizing field is small. In other words we take $N_z = 0$ for this case in Eq. (3.7a). More generally we take

$$C_0(\mathbf{k}) = g\beta H_D + 2\pi g\beta M \sin^2 \theta_k, \quad (3.8)$$

where the demagnetizing field, H_D , is assumed to be

$$H_D = 0, \quad 0 \leq H_0 \leq N_z M, \quad (3.9a)$$

$$H_D = -N_z M, \quad N_z M \leq H_0, \quad (3.9b)$$

where H_0 is the externally applied magnetic field.

Next we consider the effects of anisotropy. We will show that the anisotropy of acoustic spin waves must be of the form

$$g\beta H_A \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0(\mathbf{k}). \quad (3.10)$$

We assume the anisotropy to originate from a single-ion crystal-field Hamiltonian:

$$\mathcal{H}_A = \sum_i V_i(\mathbf{S}_i). \quad (3.11)$$

Here $V_i(\mathbf{S}_i)$ is some polynomial in the spin operators of the i th site which is consistent with the local symmetry. Again we use the substitutions of Eqs. (2.3) and (2.4) and keep only terms quadratic in the boson operators. At this stage, because of the local nature of the Hamiltonian (3.11), the coefficients in the transformed Hamiltonian are independent of wave vector. Just as for the dipolar interactions we will neglect the \mathbf{k} dependence of the coefficients one obtains when the transformation to the normal modes is made. We then conclude that the diagonal part of the Hamiltonian which refers to the acoustic spin-wave mode is of the form of Eq. (3.10). Note that terms of the type

$$K \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0^\dagger(-\mathbf{k}) \quad (3.12a)$$

are inconsistent with the over-all cubic symmetry of the lattice. The term (3.12a) corresponds to an anisotropic energy of the form

$$K(M_x^2 - M_y^2 + 2iM_x M_y), \quad (3.12b)$$

which is incompatible with the threefold symmetry about the easy, i.e., [111], axis along which the magnetization is assumed to lie. The coefficient $g\beta H_A$ can be

related to the microscopic crystal field parameters.^{26,27} Collecting the results, (3.8) and (3.10) with those of the previous section, and allowing for an externally applied magnetic field, we find the Hamiltonian for acoustic spin waves to be

$$\mathcal{H} = \epsilon(\mathbf{k}) \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0(\mathbf{k}) + D_0(\mathbf{k}) \mathbf{Q}_0^\dagger(\mathbf{k}) \mathbf{Q}_0^\dagger(-\mathbf{k}) + [D_0(\mathbf{k})]^* \mathbf{Q}_0(\mathbf{k}) \mathbf{Q}_0(-\mathbf{k}), \quad (3.13)$$

where

$$\begin{aligned} \epsilon(\mathbf{k}) = & DK^2 + EK^4 \\ & + F(K_x^2 K_y^2 + K_x^2 K_z^2 + K_y^2 K_z^2) + G(K_x^6 + K_y^6 + K_z^6) \\ & + HK^2(K_x^2 K_y^2 + K_x^2 K_z^2 + K_y^2 K_z^2) + IK_x^2 K_y^2 K_z^2 \\ & + 2\pi g\beta M \sin^2 \theta_k + g\beta H_{\text{eff}}, \end{aligned} \quad (3.14)$$

where H_{eff} is defined as

$$H_{\text{eff}} = H_0 + H_A + H_D. \quad (3.15)$$

The thermodynamic properties follow in a straightforward way¹⁴:

$$\begin{aligned} \frac{\Delta S_\nu}{S_\nu} = & \delta_\nu + \frac{2}{3} \zeta\left(\frac{3}{2}\right) \left(\frac{k_\beta T}{16\pi D}\right)^{3/2} \left\{ 1 - \frac{3}{2} \frac{1}{\zeta\left(\frac{3}{2}\right)} \left(\frac{\pi g\beta H_{\text{eff}}}{k_\beta T}\right)^{1/2} \right. \\ & - \frac{H_{\text{eff}} + 4\pi M}{2\zeta\left(\frac{3}{2}\right)} \left(\frac{g\beta}{4\pi M k_\beta T}\right)^{1/2} \sin^{-1}\left(\frac{4\pi M}{H_{\text{eff}} + 4\pi M}\right) \\ & \left. + \beta_\nu' \left(\frac{k_\beta T}{D}\right) + \gamma_\nu' \left(\frac{k_\beta T}{D}\right)^2 + \dots \right\}. \end{aligned} \quad (3.16)$$

Note that the correction proportional to $(M/T)^{1/2}$ only affects the $T^{3/2}$ term. Note also that there is an additional zero-point motion due to dipolar interactions. We have neglected this effect, since M is so much smaller than H_E . When the condition $g\beta H_{\text{eff}} \ll k_\beta T$ is not satisfied, the usual demagnetizing field treatment is more appropriate.

IV. CONCLUSION

We have calculated the magnetization of the a and d sublattices of YIG at low temperatures. The effect of dipolar interactions is significant and has been taken into account. We have neglected spin-wave interactions and have not included the effects of the optical modes. Thus the formulas given here can only be expected to be reliable below say 100°K. Although in principle our analytic expressions for the coefficients of the $T^{3/2}$, $T^{5/2}$, and $T^{7/2}$ terms in the sublattice magnetizations can be used to determine experimentally three different linear combinations of the exchange integrals, in practice only two linear combinations are essentially independent. Thus it is necessary to study the effects of the optical modes in order to determine experimentally the values of J_{aa} , J_{ad} , and

²⁶ S. Geschwind, J. Appl. Phys. **32**, 263S (1961); Phys. Rev. **121**, 363 (1961).

²⁷ L. R. Walker, J. Appl. Phys. **12**, 264S (1961).

²⁵ A. B. Harris, Phys. Rev. **143**, 353 (1966).

J_{ad} from low-temperature magnetization data. Nevertheless, our present results, when supplemented by rough estimates of the contributions from the optical modes, have been used¹² to resolve the discrepancy between Wojtowicz's analysis⁸ of the high-temperature susceptibility⁹ and studies^{10,11} of substituted garnets on the one hand, and the spin-wave analysis of low-temperature magnetization data on the other.

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APPENDIX A: ZERO-POINT MOTION IN THE PRESENCE OF SPIN-WAVE INTERACTIONS

Actually the total magnetization in the ground state, M_0 will be given by $M_{N\acute{e}el}$, its value in the Néel state, to all orders of temperature-independent perturbation theory when spin-wave interactions are taken into account. If, for example, one treats the transverse terms as a perturbation on an Ising model Hamiltonian,²⁸ the proposition is obvious, because $S_i^+ S_j^-$ commutes with $\sum_i S_i^z$. In spin-wave theory using the Dyson-Maleev transformation²⁹ the perturbation includes terms like

$$n_i a_i a_j, \quad a_i^\dagger a_j^\dagger n_j, \quad n_i n_j, \quad (A1)$$

where i refers to an a sublattice and j to a d sublattice. In the boson representation the total z component of spin is

$$\sum_i S_i^z + \sum_j S_j^z = \sum_j (S - n_j) - \sum_i (S - n_i), \quad (A2)$$

which again commutes with the perturbation terms in Eq. (A1). Similarly, using the Holstein-Primakoff transformation¹⁴ one has terms in the perturbation of the type

$$\left[\left(1 - \frac{n_i}{2S} \right)^{1/2} \left(1 - \frac{n_j}{2S} \right)^{1/2} - 1 \right] a_i a_j, \\ a_i^\dagger a_j^\dagger \left[\left(1 - \frac{n_i}{2S} \right)^{1/2} \left(1 - \frac{n_j}{2S} \right)^{1/2} - 1 \right], \quad n_i n_j. \quad (A3)$$

Again, all such terms commute with the operator in Eq. (A2).

$$\langle 0 | \mathbf{1}_d | 0 \rangle = 3, \quad (B1)$$

$$\langle 0 | \mathbf{1}_d | 1 \rangle = -\langle 1 | \mathbf{1}_d | 0 \rangle = -\sqrt{6}, \quad (B2)$$

$$\langle 1 | \mathbf{1}_d | 1 \rangle = -2, \quad (B3)$$

$$\langle n | \mathbf{1}_d | n \rangle = 0, \quad 2 \leq n \leq 4 \quad (B4)$$

$$\langle n | \mathbf{1}_d | n \rangle = 1, \quad 5 \leq n \leq 12 \quad (B5)$$

²⁸ D. L. Bullock, Phys. Rev. **137**, A1877 (1965).

²⁹ S. V. Maleev, Zh. Eksperim. i Teor. Fiz. **33**, 1010 (1957) [English transl.: Soviet Phys.—JETP **6**, 776 (1958)]; T. Oguchi, Progr. Theoret. Phys. (Kyoto) **25**, 721 (1961).

³⁰ A. Katz, Nucl. Phys. **43**, 128 (1963).

³¹ S. Geller, H. J. Williams, R. C. Sherwood, and G. P. Espinosa, J. Phys. Chem. Solids **23**, 1525 (1962).

³² G. P. Rodrigue, H. Meyer, and R. V. Jones, J. Appl. Phys. **31**, 376S (1960).

³³ F. Boutron and C. Robert, Compt. Rend. **253**, 433 (1961).

What this argument says is that perturbation terms cannot deform the wave function by the addition of amplitudes for any value of $\sum S_i^z$ which was not present in the zero-order wave function. Suppose, however, that M_0 is not equal to $M_{N\acute{e}el}$. Then the only possible way one could obtain a correct result using perturbation theory would be to do a finite-temperature calculation and then let $T \rightarrow 0$. However, as Katz³⁰ has discussed, to obtain such a crossing of energy levels one must sum over an infinite set of graphs. Clearly in this event a sophisticated calculation would be necessary.

Experimentally³¹ M_0 for YIG is found to be 10.02 Bohr magnetons per molecule as compared to $M_{N\acute{e}el}$, which is 5g Bohr magnetons per molecule, where the g value at low temperatures is³² 2.001 ± 0.002 . However, even a theory based on noninteracting spin waves would predict M_0 to differ from $M_{N\acute{e}el}$ due to anisotropy and dipolar effects. Using Geschwind's²⁶ determination of the local crystal-field parameters and Boutron and Robert's³³ evaluation of the dipolar lattice sums, one can calculate the anisotropy and Lorentz fields at each lattice site. Although these local fields are an order of magnitude larger than the average fields appearing in the acoustic spin-wave dispersion law, they are responsible for a decrease in M_0 of only about 0.01% and hence can be neglected. Thus, from a practical standpoint one may say that $M_0 = M_{N\acute{e}el}$. From a theoretical point of view it would be interesting to display a calculation of $M_0 - M_{N\acute{e}el}$ which gives a nonzero result introducing for this purpose, if need be, a nonzero external magnetic field.

APPENDIX B: MATRIX ELEMENTS REQUIRED TO EVALUATE EQS. (2.26) AND (2.28)

In Table V we give the optical mode eigenvectors and eigenvalues necessary for the evaluation of matrix elements appearing in Eqs. (2.26) and (2.28). The left eigenvectors may be obtained from the right eigenvectors using Eq. (2.13). Using these eigenvectors one can evaluate the matrix elements required. Some such matrix elements are conveniently displayed in matrix form as we have done in Tables VI and VII. The remaining necessary matrix elements are

$$\langle 0|\mathfrak{M}_6|0\rangle = (K_x^6 + K_y^6 + K_z^6) \left(\frac{-5J_{aa}}{6!256} - \frac{11J_{dd}}{4!2^{16}} + \frac{325J_{ad}}{6!2^{15}} \right) + (K_x^4 K_y^2 + K_x^4 K_z^2 + K_y^4 K_x^2 + K_y^4 K_z^2 + K_z^4 K_x^2 + K_z^4 K_y^2) \\ \times \left(\frac{-75J_{aa}}{6!256} - \frac{1575J_{dd}}{6!2^{16}} + \frac{25J_{ad}}{4!2^{15}} \right) + (K_x^2 K_y^2 K_z^2) \left(\frac{-15J_{aa}}{4!256} - \frac{5400J_{dd}}{6!2^{16}} \right), \quad (\text{B6})$$

$$\langle 0|\mathfrak{M}_4|0\rangle = \frac{1}{3 \cdot 2^{12}} \{ K^4(160J_{aa} - 85J_{ad} + 45J_{dd}) + F_1(K)(640J_{aa} + 50J_{ad} + 45J_{dd}) \}, \quad (\text{B7})$$

where

$$F_q(K) = K_x^2 K_y^2 + q K_x^2 K_z^2 + q^2 K_y^2 K_z^2, \quad q=1, \lambda; \quad (\text{B8})$$

$$\langle 1|\mathfrak{M}_4|0\rangle = \frac{5\sqrt{6}}{9 \cdot 2^{14}} \{ K^4(-192J_{aa} + 85J_{ad} - 36J_{dd}) + F_1(K)(-768J_{aa} - 50J_{ad} - 36J_{dd}) \}, \quad (\text{B9})$$

$$\langle 2|\mathfrak{M}_4|0\rangle = \frac{-25\sqrt{6}J_{ad}}{9 \cdot 2^{13}} G_1(K), \quad (\text{B10})$$

where

$$G_q(K) = K_z K_y (K_y^2 + K_z^2) + q K_x K_z (K_x^2 + K_z^2) + q^2 K_x K_y (K_x^2 + K_y^2), \quad q=1, \lambda. \quad (\text{B11})$$

$$\langle 3|\mathfrak{M}_4|0\rangle = \langle 4|\mathfrak{M}_4|0\rangle^* = \frac{-25\sqrt{6}J_{ad}}{9 \cdot 2^{12}} G_\lambda^*(K); \quad (\text{B12})$$

$$\langle 5|\mathfrak{M}_4|0\rangle = \langle 6|\mathfrak{M}_4|0\rangle^* = \frac{5\sqrt{3}}{9 \cdot 2^{14}} [\lambda^2 F_\lambda^*(K)(24J_{ad} + 18J_{dd}) + (K_z^4 + \lambda^2 K_x^4 + \lambda K_y^4)(14J_{ad} - 15J_{dd})]; \quad (\text{B13})$$

$$\langle 7|\mathfrak{M}_3|0\rangle = \frac{5\sqrt{3}iJ_{dd}}{64} K_x K_y K_z; \quad (\text{B14})$$

$$\langle 8|\mathfrak{M}_3|0\rangle = \langle 9|\mathfrak{M}_3|0\rangle = 0; \quad (\text{B15})$$

$$\langle 10|\mathfrak{M}_3|0\rangle = \frac{5\sqrt{3}i}{1536} (3J_{dd} + 4J_{ad}) [K_z(K_y^2 - K_x^2) + K_x(K_z^2 - K_y^2) + K_y(K_x^2 - K_z^2)]; \quad (\text{B16})$$

$$\langle 11|\mathfrak{M}_3|0\rangle = \langle 12|\mathfrak{M}_3|0\rangle^* = \frac{5\sqrt{3}i}{1536} (3J_{dd} + 4J_{ad}) [K_z(K_y^2 - K_x^2) + \lambda^2 K_x(K_z^2 - K_y^2) + \lambda K_y(K_x^2 - K_z^2)]. \quad (\text{B17})$$

TABLE V. Some right eigenvectors^a for $k=0$.

State	0⟩	1⟩	2⟩	3⟩	4⟩	5⟩	6⟩	7⟩	8⟩	9⟩	10⟩	11⟩	12⟩
$a1$	1	3	3	0	0	0	0	0	0	0	0	0	0
$a2$	1	3	-1	1	1	0	0	0	0	0	0	0	0
$a3$	1	3	-1	λ	λ^2	0	0	0	0	0	0	0	0
$a4$	1	3	-1	λ^2	λ	0	0	0	0	0	0	0	0
$a5$	1	3	3	0	0	0	0	0	0	0	0	0	0
$a6$	1	3	-1	1	1	0	0	0	0	0	0	0	0
$a7$	1	3	-1	λ	λ^2	0	0	0	0	0	0	0	0
$a8$	1	3	-1	λ^2	λ	0	0	0	0	0	0	0	0
$d1$	1	2	0	0	0	1	1	1	1	1	1	1	1
$d2$	1	2	0	0	0	λ	λ^2	1	λ	λ^2	1	λ	λ^2
$d3$	1	2	0	0	0	λ^2	λ	1	λ^2	λ	1	λ^2	λ
$d4$	1	2	0	0	0	1	1	1	1	1	-1	-1	-1
$d5$	1	2	0	0	0	λ	λ^2	1	λ	λ^2	-1	$-\lambda$	$-\lambda^2$
$d6$	1	2	0	0	0	λ^2	λ	1	λ^2	λ	-1	$-\lambda^2$	$-\lambda$
$d7$	1	2	0	0	0	1	1	-1	-1	-1	1	1	1
$d8$	1	2	0	0	0	λ	λ^2	-1	$-\lambda$	$-\lambda^2$	1	λ	λ^2
$d9$	1	2	0	0	0	λ^2	λ	-1	$-\lambda^2$	$-\lambda$	1	λ^2	λ
$d10$	1	2	0	0	0	1	1	-1	-1	-1	-1	-1	-1
$d11$	1	2	0	0	0	λ	λ^2	-1	$-\lambda$	$-\lambda^2$	-1	$-\lambda$	$-\lambda^2$
$d12$	1	2	0	0	0	λ^2	λ	-1	$-\lambda^2$	$-\lambda$	-1	$-\lambda^2$	$-\lambda$
N_0^a	4	24	24	6	6	12	12	12	12	12	12	12	12
E_n^b	0	$-10\mathcal{J}_{ad}$	$40J_{aa} - 30\mathcal{J}_{ad}$	$20\mathcal{J}_{ad} - 30J_{dd}$	E_7^c	$20\mathcal{J}_{ad} - 10J_{dd}$	$20\mathcal{J}_{ad} - 20J_{dd}$						

^a These vectors are unnormalized. The proper normalization is obtained by multiplying the unnormalized eigenvectors by the factor $N_0^{-1/2}$. We use the notation $\lambda = \exp(2\pi i/3)$.

^b Associated with the eigenvector $|n\rangle$ is the eigenvalue, E_n . We define $\mathcal{J}_{ad} = J_{ad} + J_{ad}'$.

^c $E_7 = 20\mathcal{J}_{ad} - 40J_{dd}$.

TABLE VI. Some matrix elements of M_1 , $h_q = K_x + qK_y + q^2K_z$ with $q=1$, λ , $C_1 = (5\sqrt{2}i/12)J_{ad}$ $C_2 = (5J_{ad}i/12)(\lambda^2 - \lambda)$.

$\begin{matrix} n \\ m \end{matrix}$	7	8	9	$\langle n M_1 m \rangle$	10	11	12
2	$-C_1h_1$	$2\lambda^2C_1h_\lambda^*$	$2\lambda C_1h_\lambda$	0	$C_1(\lambda-1)h_\lambda^*$	$C_1(\lambda^2-1)h_\lambda$	
3	$-C_1h_\lambda$	$2\lambda^2C_1h_1$	$2\lambda C_1h_\lambda^*$	$C_1(\lambda^2-\lambda)h_\lambda$	$C_1(1-\lambda)h_1$	0	
4	$-C_1h_\lambda^*$	$2\lambda^2C_1h_\lambda$	$2\lambda C_1h_1$	$C_1(\lambda-\lambda^2)h_\lambda^*$	0	$C_1(1-\lambda^2)h_1$	
5	0	0	0	λC_2h_λ	C_2h_1	$\lambda^2 C_2h_\lambda^*$	
6	0	0	0	$\lambda^2 C_2h_\lambda^*$	λC_2h_λ	C_2h_1	

Terms involving matrix elements which are not listed above or in Tables VI and VII (and whose transposes are not so listed) vanish.

TABLE VII. Some matrix elements of M_2 .^a

$\begin{matrix} n \\ m \end{matrix}$	0	1	2	$\langle n M_2 m \rangle$			5	6
				3	4			
0	$-DK^2$	C_3K^2	$-C_6f_1$	$-C_6f_\lambda$	$-C_6f_\lambda^*$	$C_{11}\lambda^2g_\lambda$	$C_{11}\lambda g_\lambda^*$	
1	$-C_3K^2$	C_4K^2	C_6f_1	C_6f_λ	$C_6f_\lambda^*$	$-C_{10}\lambda g_\lambda^*$	$-C_{10}\lambda g_\lambda$	
2	C_6f_1	C_6f_λ	$2C_8f_1$	$-C_8f_\lambda$	$-C_8f_\lambda^*$	$-C_{7\lambda}f_\lambda$	$-C_{7\lambda}f_\lambda^*$	
3	$C_6f_\lambda^*$	C_6f_λ	$-C_8f_\lambda^*$	$-C_8f_1$	$2C_8f_\lambda$	$-C_{7\lambda}f_1$	$-C_{7\lambda}f_\lambda$	
4	C_6f_λ	$C_6f_\lambda^*$	$-C_8f_\lambda$	$2C_8f_\lambda^*$	$-C_8f_1$	$-C_{7\lambda}f_\lambda^*$	$-C_{7\lambda}f_1$	
5	$C_{11}\lambda g_\lambda^*$	$C_{10}\lambda g_\lambda^*$	$C_{7\lambda}f_\lambda^*$	$C_{7\lambda}f_1$	$C_{7\lambda}f_\lambda$	C_9K^2	$-C_9\lambda g_\lambda$	
6	$C_{11}\lambda^2g_\lambda$	$C_{10}\lambda^2g_\lambda$	$C_{7\lambda}f_\lambda$	$C_{7\lambda}f_\lambda^*$	$C_{7\lambda}f_1$	$-C_9\lambda^2g_\lambda^*$	C_9K^2	

$$^a C_3 = [J_{ad}(25 - 48x - 12y)/64] \times (5\sqrt{6}/3)$$

$$C_4 = J_{ad}(-25 + 60x + 10y)/16$$

$$C_5 = 5\sqrt{6}J_{ad}/48$$

$$C_6 = 5J_{ad}/24$$

$$C_7 = 5\sqrt{2}J_{ad}/96$$

$$C_8 = 5J_{ad}/12$$

$$C_9 = 5J_{ad}/32$$

$$C_{10} = 5\sqrt{2}(J_{ad} - J_{ad})/64$$

$$C_{11} = 5\sqrt{3}J_{ad} - 2J_{ad}/192$$

$$f_q = K_y K_x + qK_x K_z + q^2 K_x K_y \text{ with } q=1, \lambda,$$

$$g_q = K_x^2 + qK_y^2 + q^2 K_z^2 \text{ with } q=1, \lambda.$$

APPENDIX C: THE SPIN-WAVE SPECTRUM OF SOME SUBSTITUTED GARNETS

In this Appendix we derive expansions for the sublattice magnetization in substituted garnets¹¹ where magnetic ions occupy, in the first case, only the d sites (e.g., $Y_3Sc_2Fe_3O_{12}$) and, in the second case, only the a sites (e.g., $Ca_3Fe_3Si_3O_{12}$). Assuming only nearest-neighbor antiferromagnetic interactions, it is easy to verify that in the classical ground state there are two oppositely magnetized sublattices. One sublattice consists of the d sites at $\tau(nd) 1 \leq n \leq 6$ and the other consists of d sites at $\tau(n'd) 7 \leq n' \leq 12$. This array has the property that a lattice site in one sublattice has nearest neighbors only in the other sublattice. The linear equations for the excitation operators of the normal modes are found exactly as for YIG. The matrix equation analogous to (2.11) is

$$\begin{pmatrix} 1D & \mathbf{X} \\ -\mathbf{X}^\dagger & -1D \end{pmatrix} \begin{pmatrix} \phi_\rho \\ \psi_\rho \end{pmatrix} = \pm \hbar\omega_\rho \begin{pmatrix} \phi_\rho \\ \psi_\rho \end{pmatrix}. \quad (C1)$$

Here $\mathbf{1}$ is the 6×6 unit matrix, ϕ_ρ and ψ_ρ are six component column vectors, $D = -20J_{ad}$, and \mathbf{X} is the 6×6 matrix formed from the right-hand six columns and

upper six rows of $-\mathfrak{M}^{dd}(-\mathbf{k})$ which was displayed in Table II. For \mathbf{k} lying along the $[111]$ direction the vectors ϕ_ρ and ψ_ρ associated with the acoustic mode are multiples of the unit vector:

$$\phi_{ac} = \alpha(k) \begin{pmatrix} 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \end{pmatrix}, \quad \psi_{ac} = \beta(k) \begin{pmatrix} 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \end{pmatrix}, \quad (C2)$$

so that Eq. (C1) reduces to

$$-4\alpha(k) + [1 + 2 \exp(ika/4) + \exp(-ika/2)]\beta(k) = \pm \frac{\hbar\omega_{ac}}{5J_{ad}} \alpha(k), \quad (C3a)$$

$$-[1 + 2 \exp(-ika/4) + \exp(ika/2)]\alpha(k) + 4\beta(k) = \pm \frac{\hbar\omega_{ac}}{5J_{ad}} \beta(k). \quad (C3b)$$

One finds for small k :

$$\hbar\omega_{ac} = (5\sqrt{2}/2)J_{ad}ak. \quad (C4)$$

Using the normalization condition,

$$|\alpha(\mathbf{k})|^2 - |\beta(\mathbf{k})|^2 = \pm 1, \quad (C5)$$

one obtains

$$\alpha(\mathbf{k}) \sim \beta(\mathbf{k}) \sim (\sqrt{2}/3)(ak)^{-1}. \quad (C6)$$

The choice of sign in the normalization condition corresponds to the twofold degeneracy of the acoustic mode of an antiferromagnet. The sublattice magnetization at low temperatures is found to be

$$\frac{\langle \Delta S \rangle}{S} = \delta + \frac{\sqrt{2}}{1125} \left(\frac{k_B T}{J_{ad}} \right)^2 + \dots \quad (C7)$$

As for YIG the effect of zero-point motion, as embodied in the term δ , is difficult to calculate, since it requires a complete calculation of the spectrum for arbitrary \mathbf{k} values.

Calculations for the case of $Ca_3Fe_3Si_3O_{12}$ are less likely to be valuable since the ordering temperature

for this compound is so low.¹¹ The calculations parallel the ones given above, so we only quote the results. For small k

$$\hbar\omega_{ac} = 10J_{aa}ak \quad (C8)$$

and the sublattice magnetization is

$$\frac{\langle \Delta S \rangle}{S} = \delta - \frac{1}{6000} \left(\frac{k_B T}{J_{aa}} \right)^2 + \dots \quad (C9)$$

Influence of the Anomalous Skin Effect on the Ferromagnetic-Resonance Linewidth in Iron*

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The ferromagnetic-resonance linewidth has been measured in iron single-crystal whiskers over a temperature range from 300 to 4.2°K at frequencies of 9.2, 22.2, and 34.8 kMc/sec. The measurements of linewidth and line shape clearly indicate the importance of nonlocal conductivity effects.

I. INTRODUCTION

THE first measurements of ferromagnetic resonance (FMR) in single-crystal iron whiskers at room temperature were carried out by Rodbell¹ at frequencies of 9 and 20 kMc/sec. The most important conclusion resulting from this work was that the linewidth in iron is dominated by the exchange-conductivity mechanism. Our recent measurements² at room temperature and frequencies of 9.2, 22.2, 34.8, and 57.8 kMc/sec confirmed Rodbell's results. In addition, with these higher frequencies, we were able to obtain a rough estimate of the Landau-Lifshitz parameter λ . However, at 20 kMc/sec, Rodbell's linewidth is some 30 Oe narrower than ours. In nickel,² we noted a similar discrepancy with Rodbell's results at low frequencies, but the linewidths agreed at the higher frequencies. Presumably, as was suggested for the nickel case, the discrepancy in iron is also due to the better surfaces of Rodbell's samples with a resulting reduced surface anisotropy. This difference is not expected to affect our conclusions.

Rodbell¹ also measured linewidths in iron below 300°K, but very few details were given apart from the comment that there was a distinct change in the character of the resonance on cooling; this he attributed to the onset of the anomalous-skin-effect region. On the other hand, the theory of Hirst and Prange³ indicated that the onset of anomalous conductivity should have no pronounced effect upon the ferromagnetic resonance,

and our preliminary measurements on iron⁴ seemed to support this prediction. In addition, we found that the observed linewidths did not increase as rapidly as expected from normal conductivity theory, in agreement with the presence of anomalous conductivity at temperatures below 77°K. The present paper is a more detailed report of our low-temperature measurements in iron, and confirms the preliminary conclusions of Ref. 4.

II. EXPERIMENTAL METHOD

The samples used were thick iron single-crystal whiskers with axes along [100] and [111]. The [100] samples and a few of the [111] samples were given to us by Professor A. V. Gold of Iowa State University, and these whiskers were used to study FMR at 22 and 35 kMc/sec. However, at lower frequencies (9 kMc/sec), because of the large magnetocrystalline anisotropy at 300°K and below, [100] whiskers could not be used, and it became necessary to grow additional [111] whiskers for this study. Although the general techniques for iron-whisker growth have been described by Brenner,⁵ a few details are included here.

The whiskers were grown by hydrogen reduction of FeCl₂ which had previously been prepared by baking FeCl₂·4H₂O in an inert atmosphere. The FeCl₂ was held in a quartz boat; quartz was used to facilitate removal of whiskers. The optimum hydrogen-flow rate was determined to be about 300 ml/min, and the optimum reduction temperature was 750°C. Furthermore, better results were obtained using hydrogen saturated with water vapor at room temperature. Only about 3% of the "raw" whiskers were found to be

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