

Spin-Wave Spectrum of Yttrium Iron Garnet*

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The spin-wave spectrum of yttrium iron garnet is treated using a Hamiltonian involving nearest-neighbor a - a , d - d , and a - d isotropic exchange interactions. Values of the exchange constants are estimated from the molecular field constants of Pauthenet. Anisotropy and magnetic dipole-dipole interactions are neglected. Twenty spin-wave modes are found, and their energies calculated at points of cubic symmetry in \mathbf{k} space. The dispersion relation of the single "acoustical" spin-wave mode is found to agree with the value previously reported by Meyer and Harris.

INTRODUCTION

A QUANTUM-MECHANICAL spin-wave treatment of the ground and low excited states in antiferromagnets has been given by Anderson,¹ using a set of approximations whereby the z components of spin operators are expanded in terms of the transverse (x, y) components. His treatment depends upon, and provides some justification for, the approximation of the antiferromagnetic ground state by a simple arrangement in which spins on different sublattices are antiparallel. This scheme was used by Kaplan in a calculation of the spin-wave spectrum of a normal spinel ferrite.² Previous semiclassical calculations had given the dispersion relations of the two modes of lowest energy.^{3,4} We have used the approximations of Anderson in calculating the spin-wave spectrum of yttrium iron garnet (YIG).

YIG has a body-centered cubic lattice. The conventional cubic unit cell contains 40 magnetic Fe^{3+} ions distributed over 24 d sites and 16 a sites. Neutron diffraction and saturation magnetization experiments show that spins on a and d sites point in opposite directions. It has been shown that, on the basis of the theory of Yafet and Kittel, canted spins should not occur in the ground state.⁵ References and a further description of the structure are given by Geller and Gilleo.⁶

There is believed to be an antiferromagnetic superexchange interaction between nearest-neighbor Fe^{3+} ions on nonequivalent sites.⁶ Antiferromagnetic coupling of neighboring Fe^{3+} ions on equivalent sites is also found experimentally.⁷ We shall assume that all interactions are isotropic, and are confined to nearest-neighbor pairs on equivalent and on nonequivalent sites.

METHOD OF CALCULATION

We assume the presence of a magnetic field in the z direction with which the spins interact through their

magnetic moments $\mathbf{u}_k = -2\mu_B \mathbf{S}_k$, and along which they are quantized. The Hamiltonian takes the form

$$\mathcal{H} = \sum_i \sum_j J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + 2\mu_B H_z \sum_k \mathbf{S}_{kz}. \quad (1)$$

Each index is summed over all spins in the crystal, so that each pair interaction occurs twice in the double sum. The exchange constant J_{ij} takes on the values J_{ad} , J_{aa} , J_{dd} , or zero, depending upon the relative locations of the i th and j th spins. Following Anderson, we approximate the z components of the spin operators as

$$S_{jz} = M_j [S - (S_{jx}^2 + S_{jy}^2 - S)/2S]. \quad (2)$$

This should be a rather good approximation for YIG, since $S = \frac{5}{2}$ is large. Only terms quadratic in spin operators are retained in the Hamiltonian. To the same degree of approximation we may use the commutation relation

$$[S_{jx}, S_{ky}] = iM_j \delta_{jk} S. \quad (3)$$

In Eqs. (2) and (3) the factor M_j is plus one if j designates an a site, minus one if j designates a d site.

Now consider the operators defined by

$$A_{k\beta}^+ = (2SN)^{-\frac{1}{2}} \sum_j e^{i\mathbf{k}\cdot\mathbf{r}_j} \xi_{\beta j} (S_{jx} + iS_{jy}), \quad (4)$$

$$[A_{k\beta}, A_{k'\beta'}^+] = \sigma_\beta \delta_{kk'} \delta_{\beta\beta'}, \quad (5)$$

and the adjoint of Eq. (4). Here \mathbf{r}_j is the position of the j th spin, \mathbf{k} is one of N wave vectors which are uniformly distributed throughout the first Brillouin zone, and N is the number of primitive unit cells in the sample. The subscript β will designate the different spin-wave modes. The coefficients $\xi_{\beta j}$ are the same for two spins which occupy identical sites in different primitive unit cells, and Eqs. (3), (4), and (5) imply the orthonormality condition,

$$\sum_j M_j \xi_{\beta j}^* \xi_{\beta' j} = -\sigma_\beta \delta_{\beta\beta'}, \quad (6)$$

where the sum is over different sites in the primitive cell. The sign factor σ_β will, according to (4) and (5), determine whether the excitation of the β th spin-wave mode increases or decreases the z component of total angular momentum.

Using relation (6), we can invert (4) to obtain the spin operators as functions of the $A_{k\beta}$ and their adjoints. Upon substitution of these functions, the Hamiltonian,

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¹ P. W. Anderson, Phys. Rev. **86**, 694 (1952).

² T. A. Kaplan, Phys. Rev. **109**, 782 (1958).

³ H. Kaplan, Phys. Rev. **86**, 121 (1952).

⁴ J. S. Kouvel, Technical Report 210, Cruft Laboratory, Harvard University, 1959 (unpublished).

⁵ P.-G. de Gennes, Phys. Rev. Letters **3**, 209 (1959).

⁶ S. Geller and M. A. Gilleo, J. Phys. Chem. Solids **3**, 30 (1957).

⁷ R. Pauthenet, Ann. phys. **3**, 424 (1958).

apart from constant terms, will take on the diagonal form

$$\mathcal{H} = \sum_{\mathbf{k}} \sum_{\beta} \frac{1}{2} [\hbar\omega_{\beta}(\mathbf{k}) + \sigma_{\beta} 2\mu_B H z] \times (A_{k\beta} A_{k\beta}^{\dagger} + A_{k\beta}^{\dagger} A_{k\beta}), \quad (7)$$

provided the $\xi_{\beta j}$ are properly chosen. The $\xi_{\beta j}$ must be components of the eigenvectors of a Hermitian matrix whose structure depends upon the spin couplings in the primitive unit cell. The dimension of this matrix is equal to the number of magnetic ions in the primitive cell; hence, we get as many modes as we have magnetic ions in the primitive cell. For a body-centered cubic lattice, the primitive cell has half the volume of the unit cube, so YIG has 20 modes. It is possible to satisfy the orthogonality requirement (6) and simultaneously to diagonalize this matrix; the solution gives the coefficients $\xi_{\beta j}$, the sign factor σ_{β} , and the frequencies $\omega_{\beta}(\mathbf{k})$. There is no guarantee that the frequencies are all positive, since the matrix is not positive definite, but the occurrence of negative frequencies would indicate a poor choice for the ground-state alignment.

In the present calculation the problem has been treated in terms of a simple cubic lattice, and the conventional cubic unit cell was used as a primitive cell. Thus, there were 40 modes, and the Brillouin zone had half the volume of the one appropriate to the bcc lattice. The same eigenvalues occur in both approaches, though half of the energy surfaces near $\mathbf{k}=0$ on the sc picture would appear near the corners of the first Brillouin zone on the bcc picture.

Since the matrix reflects the symmetries of the space group of the crystal, the problem of solution is simplified for wave vectors lying along the (111) direction of \mathbf{k} space. The eigenvectors must then transform according to irreducible representations of that part of the space group which leaves this direction unchanged.⁸ The components $\xi_{\beta i}(\mathbf{k})$ are thus restricted to such an extent that it was possible to solve the eigenvalue problem algebraically for all 40 modes at $\mathbf{k}=0$.

A formal perturbation solution shows that the energy of a mode which is not degenerate at $\mathbf{k}=0$ is expressible as a power series in components of \mathbf{k} . The cubic symmetry of the problem restricts this series to the form, $\omega_{\beta}(\mathbf{k}) = \omega_{\beta}(0) + A_{\beta} k^2 + O(k^4)$. The functional form of the energy of modes degenerate at $\mathbf{k}=0$ has not been determined.

RESULTS

The dispersion relation of the single acoustical spin-wave mode was found to be

$$\hbar\omega_1 = (5/16)(5J_{ad} - 8J_{aa} - 3J_{dd})(ka)^2 + O(ka)^4, \quad (8)$$

where $a = 12.3 \text{ \AA}$ is the lattice constant and the exchange constants are as given in the Appendix. The same result

⁸ L. P. Bouckaert, R. Smoluchowski, and E. Wigner, Phys. Rev. **50**, 58 (1936).

has been recently reported by Meyer and Harris.⁹ For $\mathbf{k}=0$ the components of the eigenvector ξ_1 are all equal and $\sigma_1 = 1$, which is consistent with the usual picture of ferromagnetic spin waves.¹⁰

The dispersion relation (8) can be simply derived by expanding the Hamiltonian (1), now taken as the energy of a system of classical spins, for small deviations from perfect spin alignment. Thus we obtain the Landau exchange stiffness A which occurs¹⁰ in the phenomenological dispersion relation $\hbar\omega_1 = 2Ak^2$ /(spin per unit volume). Using values inferred from the molecular field constants for the exchange constants, the results are

$$A = (5J_{ad} - 8J_{aa} - 3J_{dd})S^2/2a = 2.2 \times 10^{-7} \text{ erg/cm}, \quad (9)$$

$$\hbar\omega_1 = a^3 Ak^2/4S = 4.1 \times 10^{-29} k^2 \text{ erg}.$$

Experimental values of A result from measurements of the spin-wave contribution to low-temperature heat capacity. Expressed in terms of A , the results reported thus far are $2.5 \times 10^{-7} \text{ erg/cm}$,⁹ $2.7 \times 10^{-7} \text{ erg/cm}$,¹¹ and $4.3 \times 10^{-7} \text{ erg/cm}$,¹² the last value being derived from measurements made at a single temperature. The total specific heat results differ considerably from one another, and it has been suggested that the larger specific heats (and smaller A 's) are the result of magnetic impurities.^{9,12} Although present results are somewhat ambiguous, the theoretical value is at least seen to be of the correct order of magnitude, and any discrepancy is probably due to inaccuracies in values of the J 's.

Since our assumption that the a - a and d - d interactions are between nearest neighbors on equivalent sites seemed rather arbitrary, it was thought to be of interest to repeat the calculation using next-nearest neighbors. The simple classical calculation showed that the new results are obtained by multiplying the coefficients of J_{aa} or J_{dd} by 8/3 or 7/3, respectively, in formulas (8) and (9). These factors include the changes in values of exchange constants, i.e., we should still use the exchange constants given in the Appendix. On the basis of this model then, large interactions between next-nearest-neighboring spins on equivalent sites seem unlikely, since they lead to very low values of A .

The other modes have finite energy at $\mathbf{k}=0$. The simplest of these has energy $\hbar\omega_2 = 10J_{ad}$. The frequency ω_2 is equal to the exchange resonance frequency derived for a ferrite by Kaplan and Kittel using a two-sublattice model.¹³ The components of the eigenvector ξ_2 take the values ξ_{2a} and ξ_{2d} for a and d sites, respectively, where $3\xi_{2d} = 2\xi_{2a}$; and $\sigma_2 = -1$. The exchange resonance is not expected to be directly observable, due to the equality of gyromagnetic ratios of spins on a and d sites.

The remaining energy eigenvalues are given in Table

⁹ H. Meyer and A. B. Harris, J. Appl. Phys. **31**, 49S (1960).

¹⁰ C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).

¹¹ D. T. Edmunds and R. G. Peterson, Phys. Rev. Letters **2**, 499 (1959).

¹² J. E. Kunzler, L. R. Walker, and J. K. Galt, Phys. Rev. **119**, 1609 (1960).

¹³ J. Kaplan and C. Kittel, J. Chem. Phys. **21**, 760 (1953).

TABLE I. Spin-wave energy levels at the center and extreme corners of the first Brillouin zone of the bcc lattice.

n_0	n_c	σ_β	$\hbar\omega_\beta$
1	0	+	0
1	0	-	$10J_{ad}$
1	0	+	$20J_{ad}-40J_{dd}$
2	6	+	$20J_{ad}-30J_{dd}$
3	0	+	$20J_{ad}-20J_{dd}$
2	0	+	$20J_{ad}-10J_{dd}$
1	0	-	$30J_{ad}-80J_{aa}$
3	2	-	$30J_{ad}-40J_{aa}$
3	0	-	$5U^{\frac{1}{2}}+(5J_{ad}+10J_{dd}-20J_{aa})^a$
3	0	+	$5U^{\frac{1}{2}}-(5J_{ad}+10J_{dd}-20J_{aa})^a$
0	6	-	$5V^{\frac{1}{2}}+(5J_{ad}+5J_{dd}-20J_{aa})^b$
0	6	+	$5V^{\frac{1}{2}}-(5J_{ad}+5J_{dd}-20J_{aa})^b$

$$^a U = [17J_{ad}^2 - 20J_{ad}(J_{dd} + 2J_{aa}) + 4(J_{dd} + 2J_{aa})^2],$$

$$^b V = [17J_{ad}^2 - 10J_{ad}(J_{dd} + 4J_{aa}) + (J_{dd} + 4J_{aa})^2].$$

I, along with their degeneracies at the center (n_0) and corners (n_c) of the first Brillouin zone of the body-centered cubic lattice, and sign σ_β of the unit contribution of the spin wave to the z component of angular momentum of the sample. Some modes have energies which do not depend upon J_{aa} . The excitation of these modes involves distribution of a unit spin reversal over the d sites, since those components of the eigenvector which refer to a sites are zero. Corresponding modes occur in which spins on d sites are unaffected and the energy does not depend upon J_{dd} . Other modes involve excitation of spins on both a and d sites. The large numerical coefficients seem reasonable in view of the fact that the Hamilton (1) gives energies of $(30J_{ad}-40J_{aa})$ or $(20J_{ad}-20J_{dd})$ for a localized unit spin reversal on an a or d site, respectively.

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APPENDIX

R. Pauthenet finds that the paramagnetic susceptibility of YIG can be explained if we assume that in the

absence of an applied magnetic field the spins are acted upon by the following molecular fields, where \mathbf{j}_a or \mathbf{j}_d designates the magnetic moment of a mole of Fe^{3+} ions on a or d sites, respectively⁷:

$$\begin{aligned} \mathbf{H}_a &= -88\mathbf{j}_a - 6(46.6)\mathbf{j}_d; \\ \mathbf{H}_d &= -4(46.4)\mathbf{j}_a - 80\mathbf{j}_d. \end{aligned} \quad (\text{A.1})$$

Letting \mathbf{u}_i be the magnetic moment of the i th ion, we may write the energy of the system of spins in terms of these fields:

$$\mathcal{H} = -\frac{1}{2} \sum_i \mathbf{u}_i \cdot \mathbf{H}_i. \quad (\text{A.2})$$

The Hamiltonian (1) may be written in the following form:

$$\mathcal{H} = \sum_i \mathbf{u}_i \cdot \sum_j (J_{ij}/4\mu_B^2) \mathbf{u}_j. \quad (\text{A.3})$$

In Eq. (A.3) we may put $N_0\mathbf{u}_a = \mathbf{j}_a$ and $N_0\mathbf{u}_d = \mathbf{j}_d$, where N_0 designates Avogadro's number. Letting z_{ad} be the number of nearest-neighbor d sites about each a site, etc., a comparison of (A.2) with (A.3) gives

$$\begin{aligned} \mathbf{H}_a &= -(z_{aa}J_{aa}/2\mu_B^2N_0)\mathbf{j}_a - (z_{ad}J_{ad}/2\mu_B^2N_0)\mathbf{j}_d; \\ \mathbf{H}_d &= -(z_{da}J_{da}/2\mu_B^2N_0)\mathbf{j}_a - (z_{dd}J_{dd}/2\mu_B^2N_0)\mathbf{j}_d. \end{aligned} \quad (\text{A.4})$$

Comparison of (A.1) with (A.4) gives the exchange constants in terms of the molecular field coefficients. For YIG we take $z_{aa}=8$, $z_{ad}=6$, $z_{da}=z_{ad}=4$, giving

$$\begin{aligned} J_{ad} &= J_{da} = 4.8 \times 10^{-15} \text{ erg}, \\ J_{aa} &= 1.1 \times 10^{-15} \text{ erg}, \\ J_{dd} &= 2.1 \times 10^{-15} \text{ erg}. \end{aligned} \quad (\text{A.5})$$

These values agree with those quoted by Meyer and Harris.⁹

Note added in proof. After this paper had been submitted, the author became aware that a similar calculation had been performed by L. R. Walker. Walker's results, which have not been published, agree with those of Table I. However, he has derived the exchange constants from a fit to the spontaneous magnetization curve rather than the paramagnetic susceptibility, thus obtaining somewhat different values.