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Low-Lying Spectrum of Rare-Earth Iron Garnets*

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The spin-wave spectra of simplified two- and three-sublattice models of the rare-earth iron garnets are obtained, and shown to contain an iron spin-wave spectrum similar to that in ytrrium iron garnet, together with one and two optical branches, respectively. Intensities for optical excitation of $k=0$ spin waves are computed, and the effect of an external magnetic field on the resonant frequencies is also found. The consequences of anisotropic g values, which can be very different from the Landé g_J, are explored. Whereas frequencies are determined by the g values along the exchange field produced by the iron, selection rules depend on transverse g values. This results in absorption at single-ion splitting frequencies as well as at the Kaplan-Kittel exchange resonance frequency $\omega_e=\lambda(\gamma_2 M_1-\gamma_1 M_2)$. The anisotropy in the g values also produces a shift in the exchange resonance frequency, which can be related to the macroscopic anisotropy energy deduced from the anisotropic single-ion exchange splittings. Agreement with experimental data on ytterbium iron garnet, given in an accompanying paper, is excellent. Application of the theory to anisotropy effects in ferrimagnetic resonance of rare-earth-doped yttrium iron garnet is also considered. Finally, the specific heat capacity is considered and shown to be well approximated by the single-ion Shottky anomaly approximation used by Meyer and Harris.

I. INTRODUCTION

FOR a number of years yttrium iron garnet (YIG) has played a central role in research and applications concerning ferromagnetic resonance. This prominence results from its regular crystal structure, all iron ions being trivalent, so that $g=2$ and relaxation times are long. When the diamagnetic yttrium is replaced by various trivalent rare-earth ions, we again have a regular magnetic structure, but with two very different components. This difference leads to a much more complex low-lying spectrum than that of YIG, and increasing effort is now being devoted to unraveling these spectra. In addition to the purely scientific interest in understanding these systems, the splittings may have potential technical application in submillimeter masers.

The magnetization and static susceptibility of the rare-earth iron garnets have been studied in detail by Pauthenet.[1] These garnets have the formula $5\mathrm{Fe_2O_3}\cdot 3R_2\mathrm{O_3}$, where R is a trivalent rare-earth ion. The actual cubic crystallographic unit cell contains four such formula units.[2] The ferric ions are located on 16 octahedral (a) sites and 24 tetrahedral (d) sites; the rare-earth ions are on 24 (c) sites in the unit cell. Below the Curie point (~550°K), the ferric ions order into opposing sublattices a and d, with a net moment from 8 ions per unit cell or 2 per formula unit.[1] Since the g value is nearly 2, this leads to a magnetization of 10β ($\beta=$Bohr magneton) per formula unit, when the lattices are totally ordered at low temperatures. The rare-earth ions on sublattice c have an antiferromagnetic exchange coupling to the iron ions on sublattice d, tending to align their spins against the net moment of the iron ions. Since this coupling is relatively weak, the rare-earth ions order significantly only at low temperatures (<50°K). At these temperatures, the iron sublattices are essentially completely ordered. The rare-earth–rare-earth coupling is weaker still, and is neglected here. It is the purpose of this paper to discuss the low-lying excitation spectrum of this system, when it is at low temperatures so that the iron is largely ordered, and to relate this to the observable electromagnetic absorption spectra and the specific heat.

Although the many inequivalent rare-earth sites per unit cell play a key role in interpretation of the prominent anisotropy effects in these materials, one rapidly acquires a formidably complex problem if they are taken into account in detail. Hence we shall initially ignore this complexity and treat a simple model in which the rare-earth ions are all equivalent and isotropic. Similarly, we treat the iron ions as if all were on a single ferromagnetic lattice rather than being ferrimagnetically coupled. The latter simplification should actually introduce little error for the excitations of interest here. After considering some general properties of this model, we shall indicate how the complication of anisotropy and inequivalent ions can be treated, in some cases at least. Comparison with the detailed experimental results on ytterbium iron garnet (YbIG) presented in an accompanying paper[3] will illustrate the results of this analysis.

Since the microwave ferromagnetic resonance behavior of these systems has already been discussed extensively by Kittel, deGennes, and Portis,[4] and by Van Vleck,[5] that part of the spectrum will not be treated in detail here.

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* Supported in part by the U. S. Office of Naval Research, The National Science Foundation, and the Alfred P. Sloan Foundation.

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[2] F. Bertaut and F. Forrat, Compt. rend. **242**, 382 (1956).

[3] A. J. Sievers, III, and M. Tinkham, following paper, [Phys. Rev. **123**, 321 (1961)].

[4] C. Kittel, Phys. Rev. **115**, 1587 (1959); P.-G. de Gennes, C. Kittel, and A. M. Portis, Phys. Rev. **116**, 323 (1959); C. Kittel, *ibid.* **117**, 681 (1960).

[5] J. H. Van Vleck, Phys. Rev. **123**, 58 (1961).

II. ISOTROPIC TWO-SUBLATTICE MODEL

A model which gives considerable insight into the salient properties of the garnet system, yet which is simple to solve analytically, is a one-dimensional one in which there is a chain of N iron ions (1), each coupled to a rare-earth ion (2). The iron-iron ferromagnetic coupling is given by an exchange constant c, and the iron–rare-earth coupling constant is λ. We assume isotropic exchange coupling, and take $\lambda>0$ to represent antiferromagnetic coupling. We neglect the rare-earth–rare-earth coupling as negligible. If we also include an external field $\mathbf{H}$, the Bloch equations of motion are given by

$$\begin{aligned} d\mathbf{M}_{1i}/dt&=\gamma_1\mathbf{M}_{1i}\times[\mathbf{H}+c(\mathbf{M}_{1,i+1}+\mathbf{M}_{1,i-1})-\lambda\mathbf{M}_{2i}],\\ d\mathbf{M}_{2i}/dt&=\gamma_2\mathbf{M}_{2i}\times(\mathbf{H}-\lambda\mathbf{M}_{1i}), \end{aligned} \tag{1}$$

where γ_1 and γ_2 are the effective gyromagnetic ratios for the two sublattices. ($\gamma=ge/2mc$, g being the spectroscopic splitting factor, which is taken to be isotropic for the present.) We solve for small oscillations about an equilibrium configuration in which $M_{1z}=M_1$ and $M_{2z}=-M_2$ by linearizing these equations in the usual way, considering only first-order transverse components. The equations simplify if we introduce circularly polarized modes by $M_\pm=M_x\pm iM_y$. Finally, we take cognizance of the translational symmetry of the linear chain under displacements by a distance a to write for the deviations

$$\delta M_{\pm i}=\delta M_\pm e^{i(\omega t-kx_i)},$$

where the allowed values of k are given by $k_nNa=2n\pi$, with $n=0, \pm1, \pm2, \ldots, \pm(N-1)/2$, thus introducing spin-wave normal modes. If we temporarily set $\mathbf{H}=0$, we obtain the following equations of motion for normal modes:

$$\begin{aligned} \mp\omega M_{1\mp}&=-\gamma\lambda(M_2M_{1\mp}+M_1M_{2\mp})\\ &\quad-2c\gamma_1M_1(1-\cos ka)M_\mp,\\ \mp\omega M_{2\mp}&=\gamma_2\lambda(M_2M_{1\mp}+M_1M_{2\mp}). \end{aligned} \tag{2}$$

Note that changing from M_+ to M_- merely changes the sign of the frequency ω. Thus we lose no generality by considering only M_- components. Also note that if we set $\lambda=0$ to decouple the two types of ions, the normal mode frequencies become 0 for the rare-earth ions, and

$$\omega_0=\Omega_k=2c\gamma_1M_1(1-\cos ka)=4c\gamma_1M_2\sin^2(ka/2) \tag{3}$$

for the iron. This is the usual spin-wave spectrum of a ferromagnetic lattice, in which $\omega\sim k^2$ for $ka\ll1$. Introducing Ω_k into (2), we have

$$\begin{aligned} (\omega-\Omega_k-\gamma_1\lambda M_2)M_{1-}-(\gamma_1\lambda M_1)M_{2-}&=0,\\ (\gamma_2\lambda M_2)M_{1-}+(\omega+\gamma_2\lambda M_1)M_{2-}&=0. \end{aligned} \tag{4}$$

Solving the associated secular equation leads to the eigenfrequencies

$$\omega_0=\tfrac{1}{2}\{(\Omega_k-\omega_e)\pm[(\Omega_k-\omega_e)^2+4\omega_2\Omega_k]^{\frac{1}{2}}\}, \tag{5}$$

where

$$\omega_e=\lambda(\gamma_2M_1-\gamma_1M_2), \tag{6a}$$

and

$$\omega_2=\gamma_2\lambda M_1. \tag{6b}$$

In this, ω_e is the exchange resonance frequency derived by Kaplan and Kittel[6] for a two-sublattice system, and ω_2 is the frequency of precession of a single rare-earth ion in the exchange field of its iron neighbor.

The observable spectrum arises from the uniform modes ($k=0$) which can interact with a uniform applied oscillatory field. Since Ω_k goes to zero at $k=0$, Eq. (5) reduces to

$$\omega_0=0,\ -\omega_e \tag{7}$$

there. The zero-frequency mode will become the usual ferrimagnetic resonance mode in a static external magnetic field, whereas the mode at $-\omega_e$ is the exchange resonance mode of Kaplan and Kittel,[6] also discussed by Wangsness.[7]

For ka relatively large, $\Omega_k\gg\omega_e$, ω_2; and (5) reduces to

$$\omega_0\approx(\Omega_k+\omega_2-\omega_e),\ -\omega_2[1-(\omega_2-\omega_e)/\Omega_k]. \tag{8}$$

Thus over most of the spin-wave spectrum, the frequencies of the two modes are nearly equal to the unperturbed frequency of the iron spin wave Ω_k and to the single-ion splitting ω_2, respectively. The correction terms are down by a factor of order λ/c, the ratio of the exchange constants.

The entire spin-wave spectrum for this model is sketched in the upper part of Fig. 1. The magnitudes of the parameters are chosen to approximate the situation in YbIG, but the qualitative behavior would be the same for the other garnets. The significance of the dif-

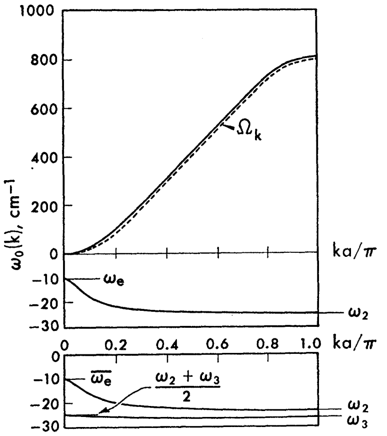

FIG. 1. Spin-wave spectra of two- and three-sublattice models of rare-earth iron garnets. Since the branch near the unperturbed iron spin-wave branch Ω_k is essentially the same in both cases, it is drawn only once. The two-sublattice case, shown above, gives one optical branch, and the three-sublattice case gives two, as shown in the lower part of the figure. Note the tenfold difference in scale between the positive and negative frequency branches.

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[6] J. Kaplan and C. Kittel, J. Chem. Phys. **21**, 760 (1953).

[7] R. K. Wangsness, Phys. Rev. **91**, 1085 (1953); **93**, 68 (1954).

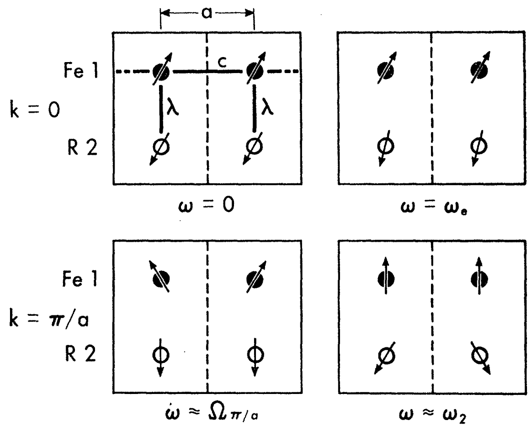

FIG. 2. Normal modes of the two-sublattice problem at $k=0$ and $k=\pi/a$. Two adjacent cells are shown in each case. The modes at $k=0$ are observable if γ_1 and γ_2 are different. The modes at $k=\pi/a$ will be observable if the transverse γ's of the ions of a given numbered type repeat only with double period $2a$, even if the longitudinal γ is the same for all ions of the type considered. For clarity, the exchange coupling network is shown only in the first case.

ference in sign of the two frequencies is that they have opposite senses of circular polarization for the case treated here.

We can get some feeling for the nature of these modes by noting from (4) that

$$\omega_0\left(\frac{M_{1-}}{\gamma_1}+\frac{M_{2-}}{\gamma_2}\right)=\Omega_k\frac{M_{1-}}{\gamma_1},$$

or

$$J_-/J_{1-}=\Omega_k/\omega_0, \tag{9}$$

where $J_-=J_{1-}+J_{2-}$. Thus when $\Omega_k\to 0$ at $k=0$, $J_-\to 0$ for the exchange resonance mode, since $\omega_e\neq 0$. Because $J_-=0$, there is a precessing moment to couple to an applied oscillating field only if there is a difference of γ_1 and γ_2, so that M_- is not also zero. The resulting absorption intensity is proportional to $(\gamma_1-\gamma_2)^2$. On the other hand, at high k values, where $\Omega_k\gg\omega_e$, we see that $J_{1-}\approx J_-$ for the mode near Ω_k and $J_{1-}\approx 0$ for the low-frequency mode. Thus, as expected, the two modes involve the iron and the rare earth separately when the frequencies are quite disparate. These modes are shown schematically in Fig. 2.

To calculate the intensity of the exchange resonance more quantitatively, we introduce an oscillating transverse field **h** in the equations of motion, and find the susceptibility for frequencies near $-\omega_e$ to be given by

$$\begin{aligned}\chi_-(\omega)&=M_-(\omega)/h_-(\omega)\\&=(\gamma_1-\gamma_2)^2/(\omega+\omega_e)[(\gamma_2/M_2)-(\gamma_1/M_1)]. \end{aligned}\tag{10}$$

The susceptibility χ_x for linear transverse polarization will be just half this value, since half the field is then circularly polarized in the wrong sense. We may compute the integrated absorption, assuming a linewidth $\Delta\omega\ll\omega_0$, by using the Kramers-Kronig relation,[8]

$$\chi_x'(\omega)=\frac{2}{\pi}\int_0^\infty\frac{\omega_1\chi_x''(\omega_1)d\omega_1}{\omega_1^2-\omega^2}, \tag{11}$$

and simplifying for ω such that $\Delta\omega\ll(\omega-\omega_0)\ll\omega_0$. In this case, (11) becomes

$$\chi_x'(\omega)=\frac{1}{\pi(\omega_0-\omega)}\int_0^\infty\chi_x''(\omega_1)d\omega_1=\frac{\chi_x''(\omega_0)\Delta\omega}{\pi(\omega_0-\omega)}. \tag{12}$$

Hence (10) implies

$$\begin{aligned}\chi_x''(\omega_0)\Delta\omega&=\frac{\pi(\gamma_1-\gamma_2)^2}{2|(\gamma_2/M_2)-(\gamma_1/M_1)|}\\&=-\frac{\pi}{2}\frac{\lambda M_1M_2(\gamma_1-\gamma_2)^2}{\omega_e},\end{aligned}\tag{13}$$

which vanishes as $(\gamma_1-\gamma_2)^2\to 0$, as expected from the structure of the normal mode. We can also use (11) to compute the contribution of the exchange resonance mode to the transverse static susceptibility. This gives

$$\chi_x'(0)=-\frac{2}{\pi}\frac{\chi_x''(\omega_0)\Delta\omega}{\omega_0}=\frac{\lambda M_1M_2(\gamma_1-\gamma_2)^2}{\omega_e^2}. \tag{14}$$

Let us now introduce a static magnetic field $\mathbf{H}_0$ directed along the z axis. Since there is no anisotropy in the model, the only stable configuration is that in which the net magnetization $(\mathbf{M}_1+\mathbf{M}_2)$ is parallel to $\mathbf{H}_0$. If we assume $M_1>M_2$, then this requires H_0 along $+z$ for the conventions assumed above. (If $M_1<M_2$, then the sign of H_0 should be reversed in the following results.) Inclusion of H_0 changes (4) to

$$\begin{aligned}(\omega-\Omega_k-\gamma_1\lambda M_2-\gamma_1H_0)M_{1-}-(\gamma_1\lambda M_1)M_{2-}&=0,\\(\gamma_2\lambda M_2)M_{1-}+(\omega+\gamma_2\lambda M_1-\gamma_2H_0)M_{2-}&=0.\end{aligned}\tag{15}$$

The resonant frequencies are now given by

$$\begin{aligned}2\omega_0=[\Omega_k-\omega_e+(\gamma_1+\gamma_2)H_0]&\\\pm\{[\Omega_k-\omega_e+(\gamma_1+\gamma_2)H_0]^2&\\+4\gamma_1\gamma_2H_0[\lambda(M_1-M_2)-H_0]&\\+4\Omega_k\gamma_2(\lambda M_1-H_0)\}^{\frac{1}{2}}.&\end{aligned}\tag{16}$$

It is of interest to specialize this to $k=0$ ($\Omega_k=0$) to get the observable resonance frequencies. In that case,

$$\begin{aligned}2\omega_0=[-\omega_e+(\gamma_1+\gamma_2)H_0]\pm[\omega_e^2-2\omega_e(\gamma_1+\gamma_2)H_0&\\+4\gamma_1\gamma_2H_0\lambda(M_1-M_2)+(\gamma_1-\gamma_2)^2H_0^2]^{\frac{1}{2}}.&\end{aligned}\tag{17}$$

Since H_0 is usually small compared to the exchange fields, it is worthwhile to expand this equation to find

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[8] See, for example, C. Kittel, *Elementary Statistical Physics* (John Wiley & Sons, Inc., New York, 1958).

the resonance frequencies to first order in H_0. These are

$$\omega_0=\frac{M_1-M_2}{(M_1/\gamma_1)-(M_2/\gamma_2)}H_0, \tag{18a}$$

$$\omega_0=-\omega_e+\frac{\gamma_2^2M_1-\gamma_1^2M_2}{\gamma_2M_1-\gamma_1M_2}H_0. \tag{18b}$$

Equation (18a) merely gives the usual ferrimagnetic resonance condition, based on the ratio of net magnetization to net angular momentum. This is the resonance discussed in detail by Kittel, deGennes, Portis, and Van Vleck[4,5] for the rare-earth iron garnets. The result (18b) gives the linear Zeeman effect for the exchange resonance. Note that it gives a shift of the resonance frequency, rather than a splitting into two components.

III. THREE-SUBLATTICE MODEL

As indicated in the Introduction, the rare-earth iron garnets actually have a very large unit cell, with many inequivalent rare-earth ions coupled to the iron lattice. Each additional ion per unit cell leads to another branch of the spin-wave spectrum, and in particular, to an additional optical mode at $k=0$. This problem rapidly becomes very cumbersome to handle, but we can illustrate the general process by adding a single additional ion per unit cell. This is taken as another rare-earth ion (3) coupled to the iron ion, but we allow $\gamma_3\neq\gamma_2$ and $\lambda_3\neq\lambda_2$. In this case, in absence of external fields, (4) is generalized to

$$\begin{aligned}&[\omega-\Omega_k-\gamma_1(\lambda_2M_2+\lambda_3M_3)]M_{1-}\\&\qquad-(\gamma_1\lambda_2M_1)M_{2-}-(\gamma_1\lambda_3M_1)M_{3-}=0,\\&(\gamma_2\lambda_2M_2)M_{1-}+[\omega+\gamma_2\lambda_2M_1]M_{2-}+0=0,\\&(\gamma_3\lambda_3M_3)M_{1-}+0+[\omega+\gamma_3\lambda_3M_1]M_{3-}=0.\end{aligned} \tag{19}$$

The associated cubic secular equation may be written as

$$\begin{aligned}\omega^3+\omega^2[\omega_{e2}+\omega_{e3}-\Omega_k]+\omega[\omega_{e2}\omega_{e3}-\omega_{13}\omega_{12}\\-(\omega_2+\omega_3)\Omega_k]-\omega_2\omega_3\Omega_k=0,\end{aligned} \tag{20}$$

where we have introduced the notation:

$$\omega_2=\gamma_2\lambda_2M_1,\quad \omega_3=\gamma_3\lambda_3M_1, \tag{20a}$$

$$\omega_{12}=\gamma_1\lambda_2M_2,\quad \omega_{13}=\gamma_1\lambda_3M_3, \tag{20b}$$

$$\begin{aligned}\omega_{e2}&=\omega_2-\omega_{12}=\lambda_2(\gamma_2M_1-\gamma_1M_2),\\ \omega_{e3}&=\omega_3-\omega_{13}=\lambda_3(\gamma_3M_1-\gamma_1M_3).\end{aligned} \tag{20c}$$

Since the general solution of the cubic is not very enlightening, we specialize immediately to the optical modes at $k=0$, where $\Omega_k=0$. From (20), we see that in this case $\omega=0$ is one root, and the secular equation reduces to a quadratic, the solutions of which are

$$\omega_0=-\left(\frac{\omega_{e2}+\omega_{e3}}{2}\right)\pm\left[\left(\frac{\omega_{e2}-\omega_{e3}}{2}\right)^2+\omega_{12}\omega_{13}\right]^{\frac{1}{2}}. \tag{21}$$

If the sublattices 2 and 3 are quite similar, it is of interest to let

$$\gamma_2=\bar{\gamma}-\delta\gamma,\quad \gamma_3=\bar{\gamma}+\delta\gamma,$$

and similarly for M and λ. If these substitutions are made in (21), first-order terms in $\delta\gamma$, etc., cancel, and we have the approximate roots,

$$\omega_0=-\bar{\gamma}\bar{\lambda}M_1, \tag{21a}$$

and

$$\omega_0=-\bar{\lambda}[\bar{\gamma}M_1-\gamma_1(2\bar{M})]. \tag{21b}$$

The root (21b) is simply the exchange resonance found for the two-sublattice model, with the combined rare-earth sublattices acting together as one sublattice. The root (21a) is a new mode whose frequency is the mean of the single-ion splittings ω_2 and ω_3. Thus we see that the nonequivalence of the two types of rare-earth ions, 2 and 3, has made possible an optical mode near the single-ion splitting frequencies, a mode which was forbidden if all rare-earth ions were completely equivalent. The fact that approximately this frequency appears also at $ka=\pi$ in the two-sublattice model can be interpreted physically as follows. When $ka=\pi$, adjacent ions on the chain move out of phase while second neighbors move in phase. Thus, if alternate ions were to have slightly different values of γ, the mode at $ka=\pi$ would also be optically active. Of course, this inequivalence amounts to doubling the size of the unit cell, producing a four-sublattice problem. If the inequivalence were very slight, one could obtain $\omega(k)$ quite accurately, within the half-size unit cell in k space, by taking the curves in Fig. 1 and folding them at $ka=\pi/2$ so that the end at $ka=\pi$ fell at $k=0$ instead.

This interpretation is confirmed by examination of the structure of the normal modes as given by the equations of motion (19). If we consider the case when $\gamma_2\approx\gamma_3$, etc., we find that the mode (21a) has $M_{2-}\approx-M_{3-}$, $M_{1-}=0$, so that the two rare earths are precessing 180° out of phase, with the iron inactive. On the other hand, the mode (21b) has $M_{2-}/\gamma_2=M_{3-}/\gamma_3=-\frac{1}{2}M_{1-}/\gamma_1$, so that the two rare earths precess in phase with each other, but out of phase with the iron, with such amplitudes that the transverse angular momentum vanishes as in the simple two-sublattice exchange resonance.

The other simple limit of the three-sublattice problem occurs at high k where $\Omega_k\gg\omega_2$, etc. Then, neglecting terms of higher order in λ/c, we obtain

$$\begin{aligned}\omega_0=(\Omega_k+\omega_{12}+\omega_{13}),\quad -\omega_2(1-\omega_{12}/\Omega_k),\\-\omega_3(1-\omega_{13}/\Omega_k).\end{aligned} \tag{22}$$

Thus, the iron spin-wave mode at Ω_k is slightly raised in frequency, as in the two-sublattice example [Eq. (8)], and we now find the single-ion splitting frequencies for both types of ion, each being decreased slightly because of the finite ratio of c/λ. (This latter correction depends on the particular coupling scheme chosen. It disappears if each rare-earth ion is assumed to be coupled equally to two adjacent iron atoms in the chain. Such a double linkage is perhaps more characteristic of the actual situation in the garnets.) Given the limits at $k=0$ and $k=\pi/a$, we can sketch the entire spin-wave spectrum of the three-sublattice system as shown in the lower part of Fig. 1. The mode near Ω_k is not redrawn since it is very similar to that in the two-sublattice case also shown in Fig. 1.

If we now repeat the argument given several paragraphs above, we may generalize our result to a six-sublattice situation in which we make adjacent cells slightly different. In that case, after folding the spectrum back at $ka=\pi/2$, we would find six frequencies at $k=0$, approximately given by 0, $\bar{\omega}_e$, $(\omega_2+\omega_3)/2$, ω_2, ω_3, $\Omega_{\pi/a}$. [More exact frequencies are given in (21) and (22).] The normal mode configurations are shown schematically in Fig. 3. Evidently the strength of the absorptions at the frequencies obtained from $ka=\pi$ will depend critically on the difference $(\gamma-\gamma')$ of the γ's in the two adjacent cells. From the argument leading to (13), we expect the intensity to vary as $(\gamma-\gamma')^2$.

When γ is not isotropic, anticipating the case considered in the following section, it is the value of γ transverse to the static polarization which governs the magnitude of the precessing moment and hence the interaction with an external oscillating field. The *frequency* of precession, however, is governed by the longitudinal value of γ along the exchange field. The folding technique described here should be quite rigorous if ions in adjacent cells differ only in transverse γ, and the intensities can be high if the difference is sizable.

To relate this model to the actual garnets, we must generalize from a one- to a three-dimensional spin-wave spectrum, and take cognizance of the full cubic unit cell containing 40 iron and 24 rare-earth ions. We do this qualitatively by taking advantage of the relative strength of the iron-iron exchange. We see from our models that the high-frequency spin waves involve essentially only the iron spins. Since the iron is relatively isotropic magnetically, we can roughly view all the iron ions as belonging to two antiparallel types; those on *a* sites and those on *d* sites. Apart from very-high-frequency modes, the 16 on *a* sites can be considered firmly locked to the 24 on *d* sites, merely changing the net moment. We can then think of the full cell being broken up into 24 subcells, each with one iron *d* site in it, and identify this subcell size with the dimension *a* in the models. The 24 rare-earth ions are coupled to *d*-site iron ions, and apart from the complication of three-dimensional geometry, it is roughly as if one were attached to each iron ion as in our two-sublattice model. Because of their anisotropy, however, there are many inequivalent types of rare-earth ions, and the unit cell must now be increased back to a size large enough to include all this variation. This increase in unit cell produces an inverse decrease in the cell in k space, accompanied by a multiplication of the number of branches of the spectrum. Alternately, we could keep the small cell in coordinate space and large cell in k space, but recognize that the inequivalence of rare-earth ions in different cells will lead to optical transitions not only at $k=0$ but also at points separated from $k=0$ by vectors of the reciprocal lattice of the full-sized coordinate unit cell. In our one-dimensional model with a full cell size of $2a$, the only example of this was at $k=\pi/a$. In the extended zone of the actual three-dimensional case, however, there would be many more such points, many with degenerate frequencies.

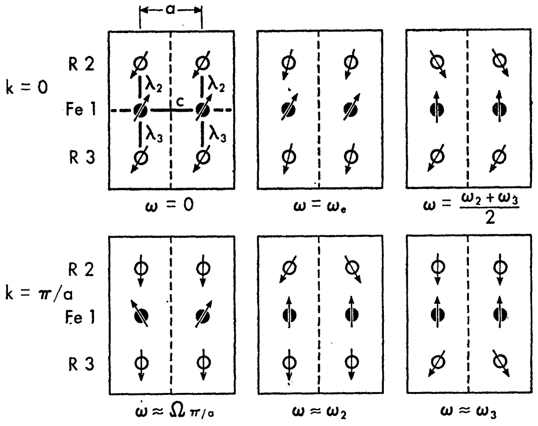

FIG. 3. Normal modes of three-sublattice problem at $k=0$ and $k=\pi/a$. Two adjacent unit cells are shown in each case. The modes at $k=0$ are optically observable, assuming γ_1, γ_2, and γ_3 are all distinct. The modes at $k=\pi/a$ will be observable if the transverse γ's of the ions of a given numbered type repeat only with double period $2a$, even if the longitudinal γ is the same for all ions of the type considered. For clarity, the exchange coupling network is shown only in the first case.

To give a concrete example of how this works, when the magnetization is along the [111] easy axis of YbIG, the rare-earth ions fall into only two classes when classified according to longitudinal γ, but there are in general several very different transverse γ's within each such class. We can view this in terms of the three-sublattice model, γ_2 and γ_3 in the model being the two different longitudinal γ's, with at least two cells having distinct transverse γ's. Alternately, we can think in terms of the two-sublattice model, with at least 4 cells being required to take account of inequivalence in both longitudinal and transverse γ. From either view, we see that the frequencies near ω_2 and ω_3 will be allowed in the spectrum, as observed experimentally.[3] If the magnetization were turned in a general direction, with only inversion symmetry left, there would be in general 12 inequivalent rare-earth ions with 6 different longitudinal γ's, and a large number of observable optical modes would exist.

IV. DISCUSSION OF PARAMETERS

g Values

In the above treatment, we have introduced *g* values or magnetomechanical ratios γ without discussion.

However, because of unquenched angular momentum, the rare-earth ions have g values far from 2, and some discussion is required. If the ions were completely unquenched, there would be no question but that one should use the usual free-ion Landé g_J factor, given by

$$g_J=\tfrac{3}{2}+[S(S+1)-L(L+1)]/2J(J+1). \tag{23}$$

However, the usual case in the garnets is that, although the spin-orbit coupling is large enough to hold the magnitude J as a good quantum number, the $(2J+1)$ different M_J states of the manifold are split apart by the crystal field with splittings[9] which are typically of the order of 100 cm^{-1}. Thus, at the low temperatures where resonance measurements must usually be made, only the lower-lying levels will be occupied. In this case, the effective g governing the occupied levels may be very different from g_J and it may be highly anisotropic.

A specific example of some importance is that the lowest level in the presence of the crystal field, but in absence of the external field or the exchange field of the iron sublattice, is a Kramers doublet. Let us review the situation here with some care. Because of the degeneracy in zero field, the proper eigenfunctions depend on field direction. Assume the field and quantization axis to lie on the z principal axis of the crystalline field, and denote the two eigenstates by ψ_a, ψ_b, which are related by time-reversal symmetry. Thus we may define

$$(a|J_z|a)=-(b|J_z|b)\equiv g_z/2g_J. \tag{24a}$$

(Throughout the paper, angular momentum is measured in units of $\hbar$, so $\mathbf{J}$ is dimensionless.) Since the magnetic interaction which these states diagonalize is $\mu_zH=g_J\beta J_zH$, the energies are $E=\pm g_z\beta H/2$, and the frequency of the transition, if allowed, is given by $h\nu=g_z\beta H$. Since J_z is diagonal in the states ψ_a and ψ_b, it follows from the angular momentum commutation relations that the diagonal matrix elements of J_x and J_y are zero. We define g_x and g_y in terms of the off-diagonal elements,

$$(a|J_x|b)=(b|J_x|a)^*=g_x/2g_J, \tag{24b}$$

$$(a|J_y|b)=(b|J_y|a)^*=ig_y/2g_J. \tag{24c}$$

If x and y are also chosen as principal axes, these g's will be real. Then for a field in an arbitrary direction, we can write

$$\mathcal{H}=g_J\beta\mathbf{J}\cdot\mathbf{H}=\beta(g_xs_xH_x+g_ys_yH_y+g_zs_zH_z), \tag{25}$$

if we take $s=\frac{1}{2}$ and identify the states $s_z=\pm\frac{1}{2}$ with the states a, b. The corresponding eigenvalues are

$$E=\pm(\beta/2)[(g_xH_x)^2+(g_yH_y)^2+(g_zH_z)^2]^{\frac{1}{2}}\equiv\pm g\beta H/2, \tag{26}$$

leading to the familiar expression for the paramagnetic resonance frequency of a Kramers doublet in a magnetic field of arbitrary orientation.[10] Thus as far as these two levels are concerned, the system acts as if the angular momentum were $s=\frac{1}{2}$ and the g values were g_x, g_y, g_z, perhaps very different from g_J. Despite this fact, the ratio of magnetic moment to true angular momentum of the ion must still be given by g_J because J remains a good quantum number. The question remains to clarify why these effective g's should be used in the Bloch equations, which on the surface are based on simple classical equations governing "true" angular momentum.

Let us conceptually simplify the problem by first considering a case with cubic but not spherical symmetry. Then $g_x=g_y=g_z=g\neq g_J$, in general. In a field H along z, the two eigenstates are separated in energy by $\Delta E=\Delta\langle\mu_z\rangle H=g_J\beta\Delta\langle J_z\rangle H=g\beta H$. Now, in the quantum description of the dynamics, the precessing transverse moment described by the Bloch equations arises from the off-diagonal matrix element of μ_x connecting ψ_a and ψ_b, which is proportional to g_x. The frequency of precession is $\omega=\Delta E/\hbar=g_z\beta H/\hbar$, which equals $g_J\beta H/\hbar=(g_Je/2mc)H$ *only* if $\Delta\langle J_z\rangle=1$. This will in fact be the case if the two eigenstates in the Kramers doublet are also eigenstates of J_z, as follows from the $\Delta J_z=\pm1$, 0 selection rules for a vector operator $\boldsymbol{\mu}$. However, the crystal field destroys the isotropy of space, and unless it is axially symmetric about H, it will in general mix several J_z values in the eigenstates of the Kramers doublet. In this case $\Delta\langle J_z\rangle=g/g_J$ can have any value up to $2J$. We require that there remain some nonzero components of ψ_a and ψ_b which satisfy the $\Delta J_z=\pm1$ selection rule only to give a precessing moment. So long as we have cubic symmetry, this condition will be met whenever $g\neq0$, since $g_x=g_y=g_z=g$.

From this discussion, we see that in treating the dynamics of these quantum systems one should use $\gamma=ge/2mc$. In order to reveal g_J when the ion is in the crystal at low temperatures, one would need to perform a gyromagnetic experiment of the Einstein-de Haas sort, in which the lattice recoil implicit in the crystal-field effect would be made observable.[11] The situation here is similar to the use of an effective mass m^* in transport theory to take account of the interaction with the periodic potential of the lattice. There, too, account must be taken of lattice momentum if the true electronic mass is to be used. The simplification resulting from use of the effective parameters is evident.

If we now relax the cubic symmetry constraint, new features appear. For one thing, we note that the *frequency* of precession of the transverse moment depends only on the g value *along* the field, while the magnitude of the transverse moment is governed by the transverse g. This should be contrasted with the classical case, where the component of the moment along the field

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[9] R. Pappalardo and D. L. Wood, J. Chem. Phys. **33**, 1734 (1960); R. L. White and J. P. Andelin, Jr., Phys. Rev. **115**, 1435 (1959).

[10] See, for example, K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 304.

[11] C. Kittel, Phys. Rev. **76**, 743 (1949); J. H. Van Vleck, *ibid.* **78**, 266 (1950).

plays no role in the torque equation. In the extreme limiting case where $g_x \rightarrow 0$, there is no oscillating moment in the x direction, and no transition could be induced by an rf field in that direction even if $g_z \neq 0$, so that a well-defined resonance frequency would exist. [When the static field is not along a principal axis, the g defined in (26) is the appropriate longitudinal g and a different expression may be derived for the transverse g values.]

This distinction between longitudinal and transverse g values is very important in determining the observable modes in the garnets. The implication is that even when the exchange field lies on a symmetry direction, such as the [111] easy axis in YbIG, where only two different splittings are observed, the number of sites with different *transverse* g values is generally much larger. Thus, modes which would be optically unobservable if the ions had isotropic g values may be quite strongly allowed with the actual anisotropic g tensor. Another way of stating this is to say that ions are equivalent only if both longitudinal and transverse g value are equal for the given applied field direction. Because this restriction is much more severe than the requirement on the longitudinal g alone, the number of inequivalent sites, and hence of observable modes, is increased markedly by anisotropy. This is the basis of the remarks at the end of Sec. III.

Exchange Field

The other parameters requiring some discussion are the exchange field parameters c and λ_i. Since the iron ions are in ^6S states, it is known that their g values are quite nearly isotropic and quite near to the spin-only value of 2. We also expect that the exchange coupling between them should be nearly isotropic. Thus, we assume that c is a simple scalar parameter. Because of the anisotropic magnetic properties of the rare-earth ions, the situation is less obvious with the parameters λ_i giving their coupling to the iron.

To get a concrete approach to the problem, let us recall that this exchange energy arises from the antisymmetry of electronic wavefunctions. In the familiar Hartree-Fock approximation, this appears as a spin-dependent potential energy term[12] arising from interaction with other electrons with net unpaired spins. This exchange potential U acting on the rare-earth ion will have a symmetry dictated by the environment, and it can be analyzed in a manner similar to the ordinary electrostatic crystal field potential V, which acts on electrons of both spins equally. The question then is: How does the magnitude of the exchange energy splitting of the rare-earth Kramers doublet depend on the orientation of the unpaired iron spins which produce the exchange field? We shall not give a detailed treatment here, but instead summarize the conclusions of an investigation along this line.

First, so long as J is a good quantum number, the net spin angular momentum in the wave function bears a fixed ratio to the total true angular momentum, namely, $\mathbf{S}=(g_J-1)\mathbf{J}$. The spin part of the moment is then $\boldsymbol{\mu}_s=[2(g_J-1)/g_J]\boldsymbol{\mu}_J$. From this, it follows that the total spin moment will show exactly the same anisotropy as $\boldsymbol{\mu}_J$ or g_{eff}. Thus, the spherically symmetric term in the exchange potential will give rise to a splitting which has the same anisotropy as the g tensor.

Second, if we take a rare-earth wave function for the case of cubic symmetry, no first-order anisotropy in the exchange splitting will result, even from noncubic terms in the exchange potential. Thus, effects of anisotropy in the exchange potential come in only in second order in terms like U_2V_2/V_c or U_2^2/V_c, where U_n, V_n are the coefficients of spherical harmonics of degree n in the exchange and crystal field potentials, respectively, and V_c represents the dominant cubic crystal-field term. Unless the exchange potential is so anisotropic that $U_2 \sim U_0$, one would expect the anisotropy from these terms to be considerably less than that coming from the g value itself. About the only relevant experimental evidence is that of Wickersheim[13] on YbIG. He finds several times as much anisotropy in the exchange splitting as in the g tensor of Yb when it is present in the homologous diamagnetic yttrium gallium and yttrium aluminum garnets. This suggests that either the exchange is actually increasing the anisotropy of the g tensor by changing the wave function or else that the exchange potential is very anisotropic, or some combination of these effects. A direct measurement of the g anisotropy in YbIG by measuring the effect of a very strong external field on the spectrum would be most helpful in resolving this question.

V. EFFECT OF ANISOTROPY ON THE SPECTRUM

In Secs. II and III we considered in detail some simple model situations characterized by isotropic g values as well as isotropic exchange. In Sec. IV we noted that inequivalence of transverse g factors was sufficient to severely affect election rules by making more ions per unit cell inequivalent even when the exchange field is along a symmetry direction. We now proceed to consider the effect of the anisotropy on the resonance frequencies themselves.

The effect on the single-ion splitting frequencies has been implicitly discussed above. For any given direction of exchange field with respect to crystal axes, there will be various splittings for the various ions per unit cell because the principal axes of their g tensors are differently oriented. For example, Wickersheim[13] has shown that in YbIG there are six different splittings for a general direction, four different splittings if the field is restricted to a (110) plane, and only two different

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[12] J. C. Slater, Phys. Rev. **81**, 385 (1951); **82**, 538 (1951); G. W. Pratt, *ibid.* **102**, 1303 (1956).

[13] K. A. Wickersheim, Phys. Rev. **122**, 1376 (1961).

splittings if the field is along a [111] or [100] direction.

The effect of the anisotropy on the exchange resonance near ω_e is more subtle. We have noted above that this mode is one in which all the rare-earth ions precess in phase with nearly equal amplitudes, despite modest differences in g. Thus, some sort of average effect of the anisotropy over all the inequivalent ions must enter. For YbIG, this problem has been attacked through direct computation by Henderson and White.[14] They have computed the exchange energy and free energy of the crystal as a function of temperature and of orientation of the exchange field with respect to crystal axes in the (110) plane. This calculation is straightforward once the exchange splittings (as a function of angle) are available input data for the statistical mechanics. Of necessity, this over-all energy has cubic symmetry and it can be fitted quite well by the usual expression,

$$F(\alpha_i,T)-F_0(T)=K_1(T)(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2)+K_2(T)(\alpha_1^2\alpha_2^2\alpha_3^2), \quad (27)$$

where α_1, α_2, α_3 are the direction cosines of the iron magnetization with respect to crystal axes. Such an expression can be used to account for the effect of anisotropy on the ferrimagnetic resonance mode, as is well known. Its application to the exchange resonance is, however, not familiar, and will be treated here.

The simplest way to introduce anisotropy into the dynamical equations is by an effective anisotropy field along the easy direction. However, with two inequivalent sublattices, it is not clear how to do this. In fact, since we known that the origin of the macroscopic anisotropy energy is in the anisotropy of the exchange splittings which depend mutually on the two sublattices, it is clear that the anisotropy field on one sublattice must be proportional to the magnetization of the other sublattice. Rather than try to force this problem into the H_A approximation, let us consider an alternate simple semiclassical model of two sublattices coupled by exchange, one sublattice being isotropic, the other having an anisotropic g value. Provided there is an easy axis of at least 3-fold symmetry (such as the [111] in YbIG), there must be axial symmetry in the average effective g tensor about this axis. That allows us to write the exchange energy as

$$E=-\lambda\gamma_1\hbar^2[\gamma_{\parallel}S_zs_z+\gamma_{\perp}(S_xs_x+S_ys_y)], \quad (28)$$

where S represents the iron and s the averaged rare earth, with $\gamma_{\parallel}\neq\gamma_{\perp}$. In this expression, $\gamma_{\perp}$ is a temperature-dependent parameter which is adjusted to reproduce the anisotropy of the macroscopic exchange energy for small angles from the easy axis (assuming λ is isotropic so that all anisotropy is in γ). Obviously, the approximation fails for large angles, because it lacks the full cubic symmetry of the problem.

To evaluate $\gamma_{\perp}$, we make contact with the macroscopic energy by letting θ_1 be the angle between $\mathbf{S}$ and the easy axis and θ_2 the angle between $\mathbf{s}$ and the opposite easy axis. Then the energy becomes

$$E=-\lambda\gamma_1\hbar^2Ss(\gamma_{\parallel}\cos\theta_1\cos\theta_2+\gamma_{\perp}\sin\theta_1\sin\theta_2). \quad (29)$$

Minimizing this with respect to θ_2 for given θ_1 leads to

$$E=-\lambda\gamma_1\hbar^2Ss(\gamma_{\parallel}^2\cos^2\theta_1+\gamma_{\perp}^2\sin^2\theta_1)^{\frac{1}{2}}, \quad (30)$$

which shows the characteristic angular dependence found in (26). Differentiating twice, we find

$$\left[-\frac{1}{E}\frac{\partial^2E}{\partial\theta^2}\right]_0=\frac{\gamma_{\parallel}^2-\gamma_{\perp}^2}{\gamma_{\parallel}^2}. \quad (31)$$

This relation allows the model parameter $\gamma_{\perp}(T)$ to be evaluated in terms of the derivatives of the free energy (27), if use is made of the fact that the torque can be computed equally well as the isothermal derivative of free energy or the isentropic derivative of the energy so that $[\partial^2E/\partial\theta^2]_0=[\partial^2F/\partial\theta^2]_0$. $E_0(T)$ is simply proportional to the product of the two sublattice magnetizations.

Now let us compute the resonant frequencies of a system with (28) as the Hamiltonian. To be certain of proper handling of this system, we use the quantum equation of motion (but with reversed sign of i to agree with our conventional $e^{+i\omega t}$ time dependence):

$$(d/dt)S_-=-(i/\hbar)(HS_--S_-H),$$

and a similar equation for s. This leads to

$$\begin{aligned}(\lambda\gamma_1\gamma_{\parallel}\hbar s_z+\omega)S_--(\lambda\gamma_1\gamma_{\perp}\hbar S_z)s_-&=0,\\ -(\lambda\gamma_1\gamma_{\perp}\hbar s_z)S_-+(\lambda\gamma_1\gamma_{\parallel}\hbar S_z+\omega)s_-&=0. \end{aligned} \quad (32)$$

Setting $\gamma_1\hbar S_z=M_1$ and $\gamma_{\parallel}\hbar s_z=-M_2$, we have the secular equation

$$\begin{vmatrix}(-\lambda\gamma_1M_2+\omega) & -\lambda\gamma_{\perp}M_1\\ \lambda\gamma_1\gamma_{\perp}M_2/\gamma_{\parallel} & (\gamma_{\parallel}M_1+\omega)\end{vmatrix}=0. \quad (33)$$

The solutions are

$$\omega_0=\frac{-\omega_e\pm[\omega_e^2+4\lambda^2M_1M_2\gamma_1(\gamma_{\parallel}^2-\gamma_{\perp}^2)/\gamma_{\parallel}]^{\frac{1}{2}}}{2} \quad (34a)$$

$$=-\frac{\omega_e}{2}\left\{1\pm\left[1+\frac{4M_1M_2\gamma_1\gamma_{\parallel}}{(\gamma_{\parallel}M_1-\gamma_1M_2)^2}\left(-\frac{1}{E}\frac{\partial^2E}{\partial\theta^2}\right)_0\right]^{\frac{1}{2}}\right\} \quad (34b)$$

$$=-\frac{\omega_e}{2}\left\{1\pm\left[1+\frac{4\gamma_1\gamma_{\parallel}\lambda}{\omega_e^2}\left(\frac{\partial^2E}{\partial\theta^2}\right)_0\right]^{\frac{1}{2}}\right\}, \quad (34c)$$

where

$$\omega_e=\lambda(\gamma_{\parallel}M_1-\gamma_1M_2), \quad (35)$$

and we have used (31) and $E_0=-\lambda M_1M_2$. We note

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

that this reduces properly to $\omega_0=0$, $-\omega_e$ when there is no anisotropy.

At an easy axis, $[\partial^2E/\partial\theta^2]_0>0$, so the anisotropy increases the exchange resonance frequency in magnitude. To calculate $\omega_0(T)$, the first approximation is to compute $\omega_e(T)$ using $M_1(T)$ and $M_2(T)$ in (35). The correction for anisotropy may then be made using (34), in which

$$\left.\frac{\partial^2E}{\partial\theta^2}\right|_0=\left.\frac{\partial^2F}{\partial\theta^2}\right|_0=-\tfrac{4}{3}[K_1(T)+\tfrac{1}{3}K_2(T)], \qquad (36)$$

where the second equality holds if the easy direction is a (111) direction. The temperature dependence is most conveniently found via (34c), but dimensional normalization is more straightforward in the form (34b). Note that since K_1 and K_2 drop much faster than M_2 as the temperature is increased, the simple approximation $\omega_0=\omega_e(T)$ becomes quite good at the higher temperatures. We note further that as the rare-earth sublattice disorders with increased temperature so that $M_2\rightarrow 0$, then $\omega_e\rightarrow\lambda\gamma_{\mathrm{II}}M_1$, the average single-ion exchange splitting for the rare-earth ions. This type of temperature dependence should be characteristic of the exchange resonance mode.

Our result (34) not only gives the effect of anisotropy on the exchange resonance, but by taking the negative sign we also obtain the frequency of the ferrimagnetic resonance mode which is raised from zero by the anisotropy. So long as the anisotropy is small, we may approximate (34c) by $\omega_0\approx(\gamma_1\gamma_{\mathrm{II}}\lambda/\omega_e)(\partial^2E/\partial\theta^2)_0$. The effect of an applied field H_0 along the easy axis will be to shift the frequency upward by the amount given by (18a). If the anisotropy is large this expression must be corrected to

$$\omega_0(H_0)=\omega_0(0)+\frac{(\lambda\gamma_1\gamma_{\mathrm{II}})(M_1-M_2)+(\gamma_1+\gamma_{\mathrm{II}})\omega_0(0)}{(\lambda\gamma_1\gamma_{\mathrm{II}})[(M_1/\gamma_1)-(M_2/\gamma_2)]+2\omega_0(0)}H_0, \qquad (37)$$

where $\omega_0(0)$ is the low-frequency root of (34).

Another application which may be noted is to the case of dilute samples of rare earth substituted into YIG.[15] In this case, the formalism should still hold, but of course M_2 is reduced in proportion to the concentration, as is $\partial^2E/\partial\theta^2$, and ω_e approaches $\gamma_{\mathrm{II}}\lambda M_1$. Therefore for low concentrations (37) takes on the limiting form

$$\omega_0(H_0)=c\frac{\gamma_1}{M_1}\left(\frac{\partial^2E^0}{\partial\theta^2}\right)_0+\gamma_1H_0\left[1+c\frac{(\gamma_2-\gamma_1)M_2{}^0}{\gamma_2M_1}\right], \qquad (38)$$

where c is the fractional concentration of rare-earth ions, and E^0 and $M_2{}^0$ represent the values for the undiluted compound. This expression gives the increase in ferrimagnetic resonance frequency (or decrease in resonance field) when H_0 lies along the easy axis. It could be generalized to an arbitrary direction, but we will not go further into that here.

VI. SPECIFIC HEAT CAPACITY

Although spectroscopic studies of the excitation spectrum give the most precise information about the garnets, the specific heat measurements of Meyer and Harris[16] have provided a most valuable initial survey which could be made independent of concern about selection rules or relaxation times. In the analysis of their experimental data, they have treated the rare-earth ions as if each had two (or more) levels split by the exchange field, leading to a Shottky anomaly in the specific heat, from which the level splitting was inferred. Dreyfus[17] has pointed out that the optical spin-wave modes at $k=0$ have energies of the same order of magnitude as these splittings, and he suggests an analysis of the specific heat in terms of optical spin waves. On the basis of our results in earlier sections, we can clarify this situation.

In Secs. II and III we noted that the complete spin-wave spectrum (shown in Fig. 1) was well approximated by an almost unmodified iron spectrum $\omega_0(k)\approx\Omega_k$ plus a number of optical branches equal to the number of rare-earth ions per unit cell. Further, beyond $ka\sim\pi/5$, these branches flatten out at nearly the single-ion splitting frequencies. Since the volume in phase space goes as k^2dk, this means that $\sim 99\%$ of the spin-wave modes will have frequencies near the splitting frequencies, while the remaining ones on the branch only are substantially lowered toward ω_e. Since ω_e is typically no less than half the single-ion frequency, even this change is not serious. Thus we conclude that the specific heat should be well accounted for by adding the contributions from the iron spin waves to the contribution from flat optical branches at the various single-ion splittings.

Since the number of spin-wave modes equals the number of ions, N, we have the following specific heat contributions for a type of ion with a single low-lying doublet with characteristic frequency ω_0, depending on whether we use the spin-wave or single-ion approximation:

$$C_{\text{s-w}}=\frac{Nk_B}{2}\frac{x^2}{\cosh x-1},$$

$$C_{\text{s-i}}=\frac{Nk_B}{2}\frac{x^2}{\cosh x+1},$$

where $x=\hbar\omega_0/k_BT$, k_B being the Boltzmann constant. As seen in Fig. 4, these agree when $k_BT\ll\hbar\omega_0$, both

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[15] J. F. Dillon, Jr., and J. W. Nielsen, Phys. Rev. **120**, 105 (1960).

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

[17] B. Dreyfus, *Proceedings of the Seventh International Conference on Low-Temperature Physics* (University of Toronto Press, Toronto, 1961), p. 127; J. Phys. Chem. Solids (to be published).

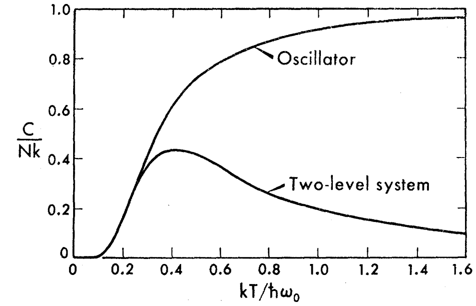

FIG. 4. Comparison of heat capacity of two-level system, representing single-ion splitting, with that of oscillator, representing spin-wave mode.

giving $Nk_Bx^2e^{-x}$. At higher temperatures they disagree, C_{s-w} approaching the constant Nk as required for an oscillator by equipartition, whereas C_{s-i} goes to zero as $(\hbar\omega_0/k_BT)^2$ when the two levels approach equal population. It is physically evident that the latter behavior is the correct one for the system at hand, since each ion can flip only once. The spin-wave approach, being linearized, fails to take this into account. Finally, we note that a completely flat spin-wave spectrum would mean no propagation. In that case, we could set up spin-wave packets which flipped a single localized spin, which would still have the same frequency ω_0. But for these wave packets it is clear that the single-ion specific heat expression is correct. Thus we conclude that the simple method of analysis used by Meyer and Harris is justified.

VII. CONCLUSIONS

We conclude that the spin-wave spectra of the rare-earth iron garnets consist of branches very similar to those in YIG together with a number of new optical branches equal to the number of magnetically inequivalent rare-earth ions in the unit cell. Inequivalence can arise from different g values transverse to the exchange field as well as from different longitudinal g values. However, approximate account can be taken of the many inequivalent rare-earth ions, if the differences are not too great (and particularly if they differ only in transverse g), by the device of neglecting differences for the purpose of finding the normal modes, but taking account of them for the purpose of determining which modes are optically observable. In this approach, one considers a small unit cell containing only a few ions, together with the correspondingly large cell in k space. Observable transitions can then occur not only at $k=0$, but at other reciprocal lattice points of the full-size unit cell. This viewpoint is closely related to the use of the extended zone scheme for approximating the energy bands of nearly free electrons.

One of the optical branches at $k=0$ will have a frequency near the exchange resonance frequency $\omega_e=\lambda(\gamma_2M_1-\gamma_1M_2)$ of Kaplan and Kittel, with an averaged rare-earth g value entering γ_2. This mode will be raised in frequency by a term proportional to the second angular derivative of the macroscopic anisotropy energy at the easy axis. As the temperature is raised, this mode will shift in frequency, both because the rare-earth magnetization M_2 drops, and because the anisotropy energy drops even faster. The other observable modes will have frequencies depending quite directly on single-ion splitting frequencies, so that they will be relatively independent of temperature. Because the bulk of the spin-wave modes lie on branches having nearly the single-ion splitting frequencies and very little dispersion, the specific heat is well approximated by the single-ion Shottky anomaly treatment used by Meyer and Harris.

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