Spin-wave spectra and thermodynamics of yttrium iron garnet—a twenty-sublattice ferrimagnet

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An approximate theory is constructed for the magnon spectra of yttrium–iron garnet, a classical object for experimental studies in magnetism. It is shown that the problem of calculating the frequencies of all 20 magnon branches over the entire Brillouin zone contains two small parameters. First, because of the large number of magnetic atoms in the unit cell the distance between nearest interacting magnetic atoms is small in comparison with the lattice constant and, accordingly, with the wavelength of a spin wave. An effective long-wavelength character thus arises in the problem. Second, there are a large number of wave-vector directions along which many elements of the Hamiltonian matrix vanish by symmetry in the basis which diagonalizes this matrix for $\mathbf{k} = 0$. These matrix elements thus have an additional, angular smallness for arbitrary directions of $\mathbf{k}$. These matrix elements can be taken into account using perturbation theory. As a result, the large elements of the Hamiltonian matrix are few in number, and they can be eliminated by several two-dimensional rotations. Approximate expressions, differing from the computer calculations by $\lesssim 10\%$, are thus obtained for the frequencies. For temperatures up to 400 K the temperature dependence of the magnetization is calculated in the spin-wave approximation, and good agreement with experiment is obtained for the following values of the exchange integrals: $J_{ad} = -35.0$ K, $J_{dd} = -14.5$ K, $J_{aa} = 0.0$ K.

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Yttrium–iron garnet ($Y_3Fe_5O_{12}$–YIG) is a marvel of nature. Its role in the physics of magnets is analogous to that of germanium in semiconductor physics, water in hydrodynamics, and quartz in crystal acoustics. There are several reasons for this. First, it has the narrowest known ferromagnetic resonance line and the lowest spin-wave damping. Second, with 80 atoms in the unit cell, each of which must find its proper location, the YIG crystal growth was so well perfected that its acoustic damping is lower than that of quartz. Third, it has a high Curie temperature $T_C = 560$ K, so that experiments can be done at room temperature. For all these reasons, YIG has become indispensable both in microwave technology and in experimental physics for studying new effects and phenomena in magnets.

Detailed studies have been made in YIG of the temperature dependence of the magnetization, specific heat, paramagnetic susceptibility, frequency and damping of long-wavelength spin waves, and much more. To analyze all these experimental data it is necessary first to know the spectrum of the elementary excitations of the magnet—the spin waves. The primitive cell of YIG contains 4 formula units of $(Y_3^{3+}Fe_2^{3+}Fe_3^{3+}O_{12}^{2-})$. Eight of the iron ions Fe^{3+} are located at octahedral (a) positions and 12 at tetrahedral (d) positions with regard to the immediate environment of O^{2-} ions.[1] There are 20 magnetic ions in all, and the magnon spectrum of YIG accordingly contains 20 branches. Their energy lies between 0 and 1000 K. In the low-temperature limit, only the long-wavelength part of the spectrum of the lower "ferromagnetic" branch is important. Its energy is easily calculated because all 20 magnetic moments in the primitive cell oscillate almost in phase and can be treated as one common magnetic moment.

In the exchange approximation[2]

$$\omega_{\mathbf{k}} = \omega_{ex}(ak)^2, \qquad \omega_{ex} = {}^5/_{16}(8J_{aa} + 3J_{dd} - 5J_{ad}), \tag{1}$$

where a is the lattice constant and ω_{ex} is the "exchange" frequency. Here J_{aa}, J_{dd}, and J_{ad} are the exchange integrals between nearest neighbors in the corresponding positions. In YIG one has $a \approx 12.5 \cdot 10^{-8}$ cm and $\omega_{ex} \approx 40$ K.

It has long been known[3] that formula (1) is valid in a rather small region of k space ($ak \lesssim 1$), with a volume less than 1% of the volume $2(2\pi/a)^3$ of the whole Brillouin zone. The magnon energy in this region does not exceed 40 K. To describe the thermodynamic and kinetic properties of YIG at higher temperatures, one cannot treat the crystal as a one-sublattice ferromagnet, even if only the lower spin-wave branch is excited. The point is that even for $ak \gtrsim 1$ one cannot assume that the magnetic moments of all 20 ions in a unit cell oscillate in phase. Therefore, to find the spin wave frequencies (including those of the lower branch) one must generally speaking solve the complete problem of the oscillations of the 20 magnetic sublattices, which amounts to the diagonalization of a 40×40 matrix. It is simple enough to calculate the frequencies of all twenty magnetic branches at $\mathbf{k} = 0$ using group theory and making use of the high degree of symmetry of the problem: the invariance of the exchange interaction with respect to rotations and the symmetry (group O_h^{10}) of the garnet lattice.[1] Harris[3] has constructed a perturbation theory for $ak \ll 1$ and found a correction $\sim k^4$ to the frequency of the lower branch (1). However, in his com-

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puter calculation he evaluated the magnon frequencies $\omega_j(\mathbf{k})$, $j = 1, ..., 20$ for the most symmetric direction $\mathbf{k}\|[111]$ and for several rather arbitrarily chosen values of the exchange integrals. It turned out that the frequencies of the 19 "optical" branches densely fill the interval from 200 to 600 K, and so generally speaking there is no temperature interval in which magnons are excited on only two branches. The widespread opinion that YIG can be treated approximately as a two-sublattice ferrimagnet is therefore lacking foundation.

In this paper we construct an approximate theory of the magnon spectra which yields analytical expressions for all 20 branches over the entire Brillouin zone (i.e., for $ak \lesssim 5$) with an acceptable accuracy of the order of 10% in a realistic interval of values of the exchange integrals. In §1, which is by way of an introduction, we give the familiar Heisenberg Hamiltonian of the problem, incorporating, as is customary,[3] the exchange interactions J_{ad}, J_{dd}, and J_{aa} between nearest neighbors. We then, for simplicity, use the quasiclassical approximation and, with the aid of a Holstein-Primakoff transformation,[4] arrive at the canonical equations of motion for the complex spin-wave amplitudes $a_j(\mathbf{k})$ and $a_j^*(\mathbf{k})$, which are the classical analogs of the Bose operators for the creation and annihilation of magnons. The quadratic part of the Hamiltonian function determines the spin-wave frequencies $\omega_j(\mathbf{k})$, $j = 1, 2, ..., 20$. To evaluate these frequencies it is generally necessary to diagonalize a 40×40 matrix. However, in the exchange approximation for a collinear structure, the problem, as we know,[3] reduces to the diagonalization of a 20×20 matrix.

Before proceeding with the analytical solution, we made detailed computer studies of the problem: using an iterative procedure we evaluated the functions $\omega_j(\mathbf{k})$ and the eigenvectors for all $j = 1, ..., 20$ different sets of exchange integrals and for the directions $\mathbf{k}\|[111]$, [100], [110], and [113]. The results of these calculations are presented in §2. Analysis of these results shows that there are hidden small parameters in the problem, which lead to the circumstance that the eigenvectors of the oscillations in a certain sense vary slowly with $\mathbf{k}$ over almost the entire Brillouin zone. One of these parameters is related to the large number of magnetic atoms in the unit cell ($N = 20$). The distance between nearest neighbors is therefore smaller than the lattice constant. As a result, a suitably constructed long-wavelength approximation is valid over the entire Brillouin zone. This is aided by the high degree of crystal symmetry, which causes a number of matrix elements to vanish in the symmetric directions [111] or [100]. Since there are many such directions, the matric elements assume a numerical "angular" smallness in arbitrary directions as well.

In §§3–5 we construct an approximate analytical theory of the magnon spectra of YIG. In §3 we transform the Hamiltonian matrix to the "irreducible" basis of the point group O_h. It is well known[5] that the representation generated by the permutation of the 8 iron ions in a positions in the unit cell of YIG decomposes into two one-dimensional representations τ_1 (identity) and τ_3 and two three-dimensional representations τ_7 and τ_9. The permutation representation of the 12 ions in d positions decomposes into two one-dimensional representations τ_1 and τ_4, two two-dimensional representations τ_5 and τ_6, and two three-dimensional representations τ_8 and τ_9. Because the representations τ_1 and τ_9 occur twice, the Hamiltonian matrix contains four pairs of off-diagonal elements even for $\mathbf{k} = 0$. In §3 we eliminate these for arbitrary $\mathbf{k}$ with the aid of four separate canonical u-v transformations, each of which involves only one pair of basis functions. In such a representation, which we call "quasinormal," the Hamiltonian matrix is diagonal for $\mathbf{k} = 0$. Its diagonal elements serve as an approximation for the spin-wave frequencies and, in the nondegenerate case, describe these frequencies well over the entire Brillouin zone. Before we given these frequencies, let us introduce the following notation for the combinations of trigonometric functions which occur in them

$$\begin{aligned} \alpha_{ijl}&=\cos 2q_i \cos q_j \cos q_l, \quad \gamma_{ijl}=\cos 2q_i \sin q_j \sin q_l,\\ \eta_{ijl}&=\cos 2q_i \cos q_j, \quad \varkappa_{ijl}=\sin 2q_i \sin q_j,\\ \nu_{ijl}&=\cos 2q_i \cos 2q_j \cos 2q_l, \quad \rho_{ijl}=\cos 2q_i \sin 2q_j \sin 2q_l. \end{aligned} \tag{2}$$

Here the indices i, j, and l take on the values $x, y, z, i\neq j\neq l\neq i$, and $\mathbf{q} = a\mathbf{k}/8$, where $\mathbf{k}$ is the wave vector of the spin waves. The functions α_{ijl}, γ_{ijl}, etc. are combined into symmetry groups $\alpha_{\pm}$, $\gamma_{\pm}$, etc., which are sums over even and odd permutations of the indices x, y, z. For example,

$$\begin{aligned} \alpha_+&={}^1/_3(\alpha_{xyz}+\alpha_{zxy}+\alpha_{yzx}), \quad \alpha_-={}^1/_3(\alpha_{zyx}+\alpha_{yxz}+\alpha_{xzy}),\\ \alpha&={}^1/_2(\alpha_++\alpha_-). \end{aligned} \tag{3}$$

The 20 branches of the magnon spectrum of YIG can be broken down into three groups according to the nature of the oscillations for $k = 0$.

1. a branches. On these branches only the spins in the a positions oscillate at $k = 0$. There are four such branches: $\omega_3(\mathbf{k}), \omega_{7j}(\mathbf{k}), j = 1, 2, 3$. Here the first frequency index corresponds to the number of the irreducible representation, and the second to the number of the basis function in the irreducible representation, if it is not one-dimensional. The frequencies of the a branches in the quasinormal approximation are:

$$\begin{aligned} \omega_3^0(\mathbf{k})&=\omega_a+40J_{aa}\nu, \quad \omega_a=-30J_{ad}+40J_{aa},\\ \omega_{71}^0(\mathbf{k})&=\omega_a+40J_{aa}\rho, \quad \omega_{72}^0(\mathbf{k})=\omega_{73}^0(\mathbf{k})=\omega_a-20J_{aa}\rho. \end{aligned} \tag{4}$$

2. d branches. On these branches only the d spins oscillate at $\mathbf{k} = 0$:

$$\begin{aligned} &\omega_4^0(\mathbf{k})=\omega_d+20J_{dd}\alpha, \quad \omega_d=-20(J_{ad}-J_{dd}),\\ &\omega_{51}^0(\mathbf{k})=\omega_{52}^0(\mathbf{k})=\omega_d+10J_{dd}\alpha, \omega_{61}^0(\mathbf{k})=\omega_{62}^0(\mathbf{k})=\omega_d-10J_{dd}\alpha,\\ &\omega_{81}^0(\mathbf{k})=\omega_d+20J_{dd}\gamma, \quad \omega_{82}^0(\mathbf{k})=\omega_{83}^0(\mathbf{k})=\omega_d-10J_{dd}\gamma. \end{aligned} \tag{5}$$

The notation here follows the same principles as for the a branches.

3. a–d branches. Here the spins at both the a and d positions oscillate. These branches arise because the same irreducible representations (τ_1 and τ_9) occur in the decomposition of the a and d permutation representations. Since the identity representation τ_1 is one-dimensional, while the representation τ_9 is three-dimensional, there are eight a–d branches. Their frequencies in the quasinormal approxima-

tion can be written in the form

$$\left.\begin{matrix}\omega_{ap}(\mathbf{k})\\ \omega_{dp}(\mathbf{k})\end{matrix}\right\} = {}^1/_2\{[(A_p+D_p)^2-4|B_p|^2]^{1/2}\pm(A_p-D_p)\}; \quad (6)$$

$$p=1,\ 91,\ 92,\ 93,\quad A_1(\mathbf{k})=\omega_a-40J_{aa}\nu,$$
$$A_{91}=\omega_a-40J_{aa}\rho,\quad A_{92}=A_{93}=\omega_a+20J_{aa}\rho,$$
$$D_1=\omega_d-20J_{dd}\alpha,\quad D_{91}=\omega_d-20J_{dd}\gamma, \quad (7)$$
$$D_{92}=D_{93}=\omega_d+10J_{dd}\gamma,\quad B_1=10\sqrt{6}J_{ad}\eta,$$
$$|B_{91}|=10\sqrt{2}|J_{ad}|(\eta-\varkappa),\quad |B_{92}|=|B_{93}|=10\sqrt{2}|J_{ad}|(\eta-\varkappa/2).$$

The letter index d is assigned to four modes [including the ferromagnetic (FM) mode τ_{d1}]. The excitation of a magnon of this type decreases the magnetization of YIG by one Bohr magneton, just as the excitation of any d-branch magnon does. On the other hand, the excitation of a magnon of the four branches with index a [including the antiferromagnetic (AFM) mode τ_{a1}] increases the magnetization by one Bohr magneton. This property is shared by any a mode.

In §4 we obtain corrections to expressions (4), (5), and (6) for the frequencies $\omega^0_{d1}(\mathbf{k})$, $\omega^0_{a1}(\mathbf{k})$, $\omega^0_4(\mathbf{k})$ and $\omega^0_3(\mathbf{k})$ of the spin waves which are nondegenerate at $\mathbf{k}=0$, using second-order perturbation theory in the off-diagonal elements of the Hamiltonian matrix in the quasinormal representation. The expressions obtained for the frequencies are entirely satisfactory and differ from the results of the numerical calculation by no more than a few percent.

In §5 we construct a perturbation theory for the frequencies of the spin waves which are doubly and triply degenerate at $\mathbf{k}=0$. The problem here is complicated by two factors. First, because of the degeneracy it is necessary to exactly diagonalize matrices of dimensionality 2×2 or 3×3. Second, the series expansion in $\mathbf{k}$ of several matrix elements which "mix" nearby modes of different representations begins with the first power of $\mathbf{k}$. Therefore, in a significant part of the Brillouin zone these matrix elements cannot be taken into account by perturbation theory and one is obliged to make exact canonical transformations to eliminate them. Only in this representation can one use perturbation theory. We have carried out this program and found the frequencies of the 20 spin-wave branches for the symmetric directions $\mathbf{k}\|[100]$ and $\mathbf{k}\|[111]$. Then, for arbitrary directions of $\mathbf{k}$ we study qualitatively the frequencies of the degenerate spin-wave branches, which have activation energies of no more than 300–400 K. Finally, in §6 we calculate the temperature dependence of the magnetization and specific heat of YIG in the approximation of noninteracting spin waves. By comparing our results with the experimental data on $M(T)$, we refine the values of the exchange integrals. We find that for

$$J_{ad}=-35.0\text{ K},\quad J_{dd}=-14.5\text{ K},\quad J_{aa}=0\text{ K} \quad (8)$$

the experimental and theoretical functions $M(T)$ agree to within the experimental accuracy ($\sim10\%$) in the interval from 0 to 400 K.

§1. QUADRATIC HAMILTONIAN AND DIAGONALIZATION PROCEDURE

We write the Heisenberg Hamiltonian for the spin system:

$$\mathcal{H}=-2\sum_n\Big\{J_{aa}\sum_{i=1,\,j>i}^{8}\mathbf{S}_i(\mathbf{R}_{in})\sum_{\mathbf{d}_{ij}}\mathbf{S}_j(\mathbf{R}_{in}+\mathbf{d}_{ij})$$
$$+J_{ad}\sum_{i=1}^{8}\sum_{j=9}^{20}\mathbf{S}_i(\mathbf{R}_{in})$$
$$\times\sum_{\mathbf{d}_{ij}}\mathbf{S}_j(\mathbf{R}_{in}+\mathbf{d}_{ij})+J_{dd}\sum_{i=9,\,j>i}^{20}\mathbf{S}_i(\mathbf{R}_{in})\sum_{\mathbf{d}_{ij}}\mathbf{S}_j(\mathbf{R}_{in}+\mathbf{d}_{ij})\Big\}. \quad (1.1)$$

Here n is the number of the primitive cell, i and j are the numbers of the sublattices: $i=1,\ldots,8$ for a ions, $i=9,\ldots,20$ for d ions, $\mathbf{S}_i(\mathbf{R}_{in})$ are the spin and coordinate of the i-th sublattice in the n-th cell, and $\mathbf{d}_{ij}$ is the distance between nearest neighbors of the i-th and j-th sublattices.

Assuming the motion of the spins to be quasiclassical, we go over from the spin variables $\mathbf{S}$ at each lattice site to the complex canonical variables a and a^* with the aid of the classical analog of the Holstein-Primakoff transformation. To determine the spectrum and eigenvectors of the oscillations, in this paper we consider the quadratic part of the Hamiltonian ($\mathcal{H}^{(2)}$) and transform into $\mathbf{k}$ space by the formula

$$a_j(\mathbf{k})=\frac{1}{N^{1/2}}\sum_n a_{jn}\exp(-i\mathbf{k}\mathbf{R}_{jn}), \quad (1.2)$$

where N is the number of cells in the crystal. Then

$$\mathcal{H}^{(2)}=\sum_{\mathbf{k}}\mathcal{H}_{\mathbf{k}},\quad \mathcal{H}_{\mathbf{k}}=\sum_{i,j=1}^{20}\{F_{ij}a_i^*a_j+[G_{ij}a_i^*(\mathbf{k})a_j^*(-\mathbf{k})+\text{c.c.}]\}$$
$$=\sum_{i,j=1}^{8}A_{ij}(\mathbf{k})a_i^*(\mathbf{k})a_j(\mathbf{k})+\sum_{i,j=9}^{20}D_{ij}(\mathbf{k})a_i^*(\mathbf{k})a_j(\mathbf{k})$$
$$+\sum_{i=1}^{8}\sum_{j=9}^{20}[B_{ij}(\mathbf{k})a_i^*(\mathbf{k})a_j^*(-\mathbf{k})+\text{c.c.}], \quad (1.3)$$

$$A_{ij}=A\delta_{ij}+2S_0J_{aa}\gamma_{ij}(\mathbf{k}),\quad A=2S_0(6J_{ad}-8J_{aa}),$$
$$D_{ij}=D\delta_{ij}+2S_0J_{dd}\gamma_{ij}(\mathbf{k}),\quad D=8S_0(J_{ad}-J_{dd}),$$
$$B_{ij}=2S_0J_{ad}\gamma_{ij}(\mathbf{k}),\quad \gamma_{ij}(\mathbf{k})=\sum_{\mathbf{d}_{ij}}\exp(i\mathbf{k}\mathbf{d}_{ij}),\quad A_{ji}=A_{ij}^*,\ D_{ji}=D_{ij}^*. \quad (1.4)$$

A similar form of the matrices A, B, and C is given in the paper by Harris,[3] from which we have adopted the notation in (1.4). The appearance of blocks of zeros in the matrices F_{ij} and G_{ij} is a consequence of the collinearity of the sublattices and the high degree of symmetry of the exchange interaction.

As a result, the problem of finding the eigenfunctions and eigenvectors of the spin-wave oscillations reduces to the diagonalization of a 20×20 matrix

$$\Lambda=\begin{pmatrix}A & B\\ -B^+ & D\end{pmatrix}$$

by means of a transformation to the new variables $b_j(\mathbf{k})$:

$$a_i(\mathbf{k}) = \sum_{j=1}^{8} u_{ji}^{(1)*}(\mathbf{k})\, b_j(\mathbf{k}) - \sum_{j=9}^{20} v_{ji}^{*}(\mathbf{k})\, b_j^{*}(-\mathbf{k}), \quad i=1,\ldots,8,$$
$$a_i^{*}(-\mathbf{k}) = -\sum_{j=1}^{8} v_{ji}^{*}(\mathbf{k})\, b_j(\mathbf{k}) + \sum_{j=9}^{20} u_{ji}^{(2)*}(\mathbf{k})\, b_j^{*}(-\mathbf{k}), \quad i=9,\ldots,20, \tag{1.5}$$

in which the Hamiltonian assumes the form

$$\mathcal{H}_{\mathbf{k}} = \sum_{j=1}^{20} \omega_j(\mathbf{k})\, b_j^{*}(\mathbf{k})\, b_j(\mathbf{k}). \tag{1.6}$$

The first eight eigenvalues Λ here coincide with the eigenfrequencies, while the remaining twelve differ in sign.

In order to make concrete computer calculations, one must assign numerical values to the exchange integrals J_{ad}, J_{dd}, and J_{aa}. At the present time, only the "intersublattice" exchange in YIG is well enough known: $J_{ad} = -(33$–$36)$ K, according to the data of different authors.[6,7] To make direct use of the experimental results on a–a and d–d exchange is difficult, however, since the values obtained for the constants J_{aa} and J_{dd} depend strongly on the particular physical effects and data-processing methods used. The opinion that $|J_{aa}| \ll |J_{dd}|$ seems rather plausible to us, since the distance between nearest a positions ($\sqrt{3}a/4$) is $\sqrt{2}$ times greater than the distance between d positions, and the exchange interaction falls off exponentially with distance. The relation $|J_{aa}/J_{dd}| \ll 1$ is also confirmed by our analysis of the temperature dependence of the magnetization as a function of exchange constants [see (8)]. It must be said that some of the data obtained by the molecular-field method imply that J_{aa} is comparable in size to J_{dd}. We attribute this disagreement to the inadequacy of the molecular-field approximation (se Ref. 6), and we shall assume that J_{aa} is negligibly small.

To evaluate the magnon spectrum we need the values of the exchange integrals, and for this we shall use the experimental data on the spectrum of the long-wavelength spin waves of the ferromagnetic branch. The value of ω_{ex} has been accurately measured in experiment and is expressed quite simply in terms of the exchange integrals in (1), thereby yielding the relations between these integrals.

Assuming in accordance with what was said above that $J_{ad} = -35$ K and $J_{aa} = 0$, we obtain the desired set of values

$$J_{dd}=-16 \text{ K}, \quad J_{ad}=-35 \text{ K}, \quad J_{aa}=0. \tag{1.7}$$

We shall call these the "standard" values of the exchange integrals. As the "admissible" limits of their variation we adopt

$$J_{aa}=-(0-3) \text{ K}, \quad J_{dd}=-(16\pm3) \text{ K}, \quad J_{ad}=-(35\pm3) \text{ K} \tag{1.8}$$

with the understanding that the combination ω_{ex} [Eq. (1)] remain equal to 41 K.

§2. SPIN-WAVE SPECTRUM AND THE NORMAL VARIABLES OF THE QUADRATIC HAMILTONIAN—THE RESULTS OF COMPUTER CALCULATIONS

2.1. The magnon spectrum evaluated by computer for $\mathbf{k}\|[100]$, [111], [110], and [113] with the standard values (1.7) of the exchange integrals is given in Figs. 1–3. The behavior of the ferromagnetic mode $\omega_{d1}(\mathbf{k})$ at small values of

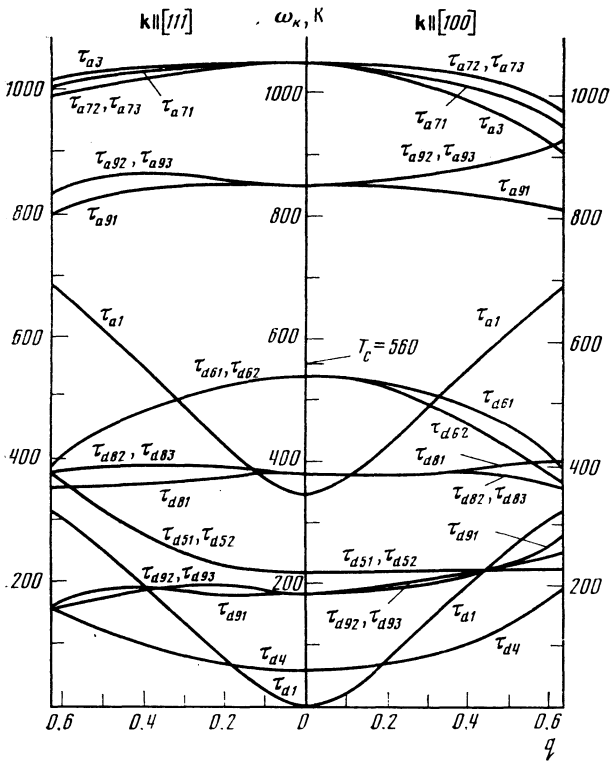

FIG. 1. Spin-wave spectrum for the symmetric directions $\mathbf{k}\|[100]$, [111] and for the standard values of the exchange integrals; $g = ak/8$.

ak is, of course, described by formula (1).

An interesting question is, to what values of ak does the dispersion relation remain quadratic to an accuracy of, say, 10%? The answer can be obtained by analyzing the next term of the expansion in (1). For the standard values of J [Eq. (1.7)] the coefficient of the $(ak)^4$ term is 8.5 K, and so the sought value is $ak \simeq 0.7$. For such values of k, we find $\omega_{d1} = 25$ K. At larger values of the frequency $\omega_{d1}(\mathbf{k})$ the dispersion relation is not in any sense quadratic. It is of fundamental importance that as ak increases the dispersion relation approaches a straight line.

2.2. In an overview of the spectra it strikes one that the

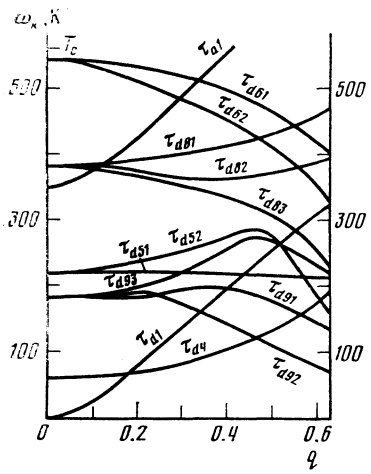

FIG. 2. Low-frequency part of the spectrum for $\mathbf{k}\|[110]$.

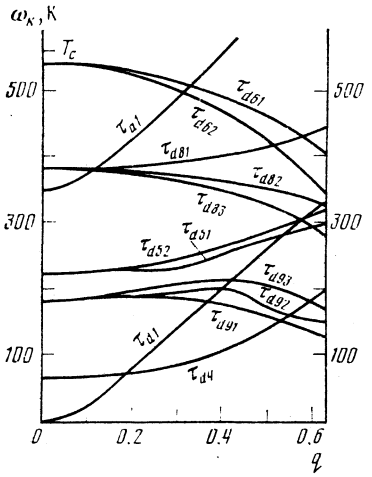

FIG. 3. Low-frequency part of the spectrum for $\mathbf{k}\|[113]$.

antiferromagnetic branch runs almost parallel to the ferromagnetic branch and is not noticeably perturbed in its multiple crossing of other branches. In the language of perturbation theory this means that the eigenvectors of the FM and AFM modes are practically unmixed with the other eigenvectors. Neglecting such intermixing, we can obtain simple analytical expressions for the frequencies $\omega_{a1}(\mathbf{k})$ and $\omega_{d1}(\mathbf{k})$ of the FM and AFM branches over the entire Brillouin zone. To do this we assume that in (1.3) the oscillation amplitudes of all eight a and all twelve d spins are equal ($a_1 = \ldots = a_8$, $a_9 = \ldots = a_{20}$) and obtain

$$i\frac{da_i}{dt} = A_1 a_i + B_1 a_j^*, \qquad -i\frac{da_j^*}{dt} = B_1^* a_i + D_1 a_j^*,$$

$$i = 1, \ldots, 8, \qquad j = 9, \ldots, 20. \tag{2.1}$$

Hence, we obtain for $\omega_{d1}(\mathbf{k})$ and $\omega_{a1}(\mathbf{k})$ the expressions (6) given in the Introduction. The notation used for the coefficients is given in (7).

Our assumption that the amplitudes of the oscillations of the spins of the a and d ions are equal is equivalent to the replacement of the 20-sublattice ferrite by a two-sublattice model. Here, however, we have assumed that the phases of the oscillations of spins a and d are not the same over the unit cell, but rather $a(\mathbf{R}_{jn}) \sim \exp(i\mathbf{k}\cdot\mathbf{R}_{jn})$, in accordance with (1.2). The presence of a phase factor is a fundamental improvement over the naive two-sublattice model with identical phases of the oscillations within a unit cell. The two-sublattice approximation proposed here, as we shall show in §4, gives a good quantitative description of the ferromagnetic and "antiferromagnetic" magnons over almost the entire Brillouin zone.

This success permitted us to suppose that the intermixing of all the remaining modes at values of ak that are not small is nevertheless in some sense small. To demonstrate this we determined in a numerical experiment the projections of the 20 eigenvectors for $\mathbf{k} \neq 0$ onto the eigenbasis for $\mathbf{k} = 0$ and found that the transition matrix between these bases is nearly block diagonal, with the eigenvectors of the same representations intermixed in the blocks.

§3. QUADRATIC HAMILTONIAN FOR SPIN WAVES IN QUASINORMAL VARIABLES

Making use of the experience of previous investigators in regard to the application of group theory for analysis of the spectrum of homogeneous oscillations ($\mathbf{k} = 0$) of the magnetic system of YIG,[1-3] let us formulate an analytical theory for evaluating the frequencies $\omega_j(\mathbf{k})$ of all 20 types of spin waves.

3.1. Our goal in this section, which is of a preparatory nature, is to write the quadratic Hamiltonian $\mathscr{H}_{\mathbf{k}}$ (for any $\mathbf{k}$) in the irreducible (for $\mathbf{k} = 0$) representation [i.e., after the transformation to the new basis by formula (1.5)], in which

$$u_{ij}^{(1)} = u_{aij}, \qquad u_{ij}^{(2)} = u_{dij}, \qquad v_{ij} = 0. \tag{3.1}$$

The matrices U_a and U_d are given in Ref. 8. The quadratic Hamiltonian is specified by the matrices $A^{(1)}(\mathbf{k})$, $D^{(1)}(\mathbf{k})$, and $B^{(1)}(\mathbf{k})$ with the aid of the relations

$$A^{(1)}(\mathbf{k}) = U_a A(\mathbf{k}) U_a^{-1}, \quad D^{(1)}(\mathbf{k}) = U_d D(\mathbf{k}) U_d^{-1}, \quad B^{(1)}(\mathbf{k}) = U_a B(\mathbf{k}) U_d^{-1}. \tag{3.2}$$

In writing the results of the matrix multiplication, we shall assign double indices to the rows and columns: $A^{(1)}_{mj,nl}$, $D^{(1)}_{mj,nl}$, and $B^{(1)}_{mj,nl}$. The first indices m and n give the irreducible representations, and i and j give the number of the basis function within a single representation, if it is not one-dimensional. Further, we shall separate out the diagonal terms in the matrices $A^{(1)}$ and $D^{(1)}$ and reduce the remaining terms to dimensionless form

$$A^{(1)}_{mj,nl} = \omega_a \delta_{m,n}\delta_{j,l} + 20 J_{aa} a_{mj,nl},$$

$$D^{(1)}_{mj,nl} = \omega_d \delta_{m,n}\delta_{j,l} + 10 J_{dd} d_{mj,nl}, \tag{3.3}$$

$$B^{(1)}_{mj,nl} = -10\sqrt{6} J_{ad} b_{mj,nl}.$$

In this normalization the maximum values of the matrix elements a, b, and d which do not vanish as $\mathbf{k} \to 0$ are equal to unity. The matrices we seek are expressed in terms of combinations of trigonometric functions. Some of these combinations, α, γ, η, $\varkappa$, ρ, and ν were already defined by formulas (2) and (3) in the Introduction. The remaining combinations are:

$$\begin{aligned} \beta_{ijl} &= \sin 2q_i \sin q_j \sin q_l, \quad & \delta_{ijl} &= \sin 2q_i \cos q_j \cos q_l, \\ \varepsilon_{ijl} &= \cos 2q_i \sin q_j \cos q_l, \quad & \zeta_{ijk} &= \sin 2q_i \cos q_j \sin q_l, \\ \theta_{ijl} &= \cos 2q_i \sin q_j, \quad & \mu_{ijl} &= \sin 2q_i \cos q_j. \end{aligned} \tag{3.4}$$

Here, as before, the indices i, j, and l take on the values x, y, and z, $\mathbf{q} = a\mathbf{k}/8$, and $i \neq j \neq l \neq i$. Functions (3.4), like functions (2), are combined into symmetric combinations $\beta_{\pm}$, $\delta_{\pm}$, ... according to rule (3). In addition, a new type of symmetric combinations arises: $\tilde{\alpha}_{\pm}$, $\tilde{\beta}_{\pm}$, $\tilde{\gamma}_{\pm}$, etc.—the sums over the even and odd permutations of x, y, and z with weight factors of 1, $\lambda = \exp(2i\pi/3)$, and λ^2. For example,

$$\tilde{\alpha}_{+} = {}^1/_3(\lambda^2\alpha_{xyz} + \lambda\alpha_{zxy} + \alpha_{yzx}),$$
$$\tilde{\alpha}_{-} = {}^1/_3(\lambda^2\alpha_{zyx} + \lambda\alpha_{yxz} + \alpha_{xzy}), \quad \tilde{\alpha} = {}^1/_2(\tilde{\alpha}_{+} + \tilde{\alpha}_{-}). \tag{3.5}$$

The remaining definitions ($\tilde{\beta}_{\pm}$, $\tilde{\gamma}_{\pm}$, etc.) differ only by replacement of the Greek letter.

To describe the Hamiltonian matrix in the irreducible basis we begin with the most awkward matrix D. Its diagonal

blocks with respect to the irreducible representations are of the form

$$d_{11}=-d_{44}=2\alpha(\mathbf{k}),$$

$$d_{66}=-d_{55}=\begin{pmatrix}\alpha & -\tilde{\alpha}^*\\ -\tilde{\alpha}^* & \alpha\end{pmatrix},\quad d_{88}=d_{99}=\begin{pmatrix}-2\gamma & \tilde{\gamma}^* & \tilde{\gamma}\\ \tilde{\gamma} & \gamma & -2\tilde{\gamma}^*\\ \tilde{\gamma}^* & -2\tilde{\gamma} & \gamma\end{pmatrix}, \tag{3.6}$$

$$d_{14}=d_{41}^*=2i\beta,\quad d_{46}=-d_{15}=(\tilde{\alpha}^*,\tilde{\alpha}),\quad d_{45}=-d_{16}=i(\tilde{\beta}^*,\tilde{\beta}),$$

$$d_{56}=i\begin{pmatrix}\beta & -\tilde{\beta}\\ -\tilde{\beta}^* & \beta\end{pmatrix},\quad d_{89}=i\begin{pmatrix}-2\delta & \tilde{\delta}^* & \tilde{\delta}\\ \tilde{\delta} & \delta & -2\tilde{\delta}^*\\ \tilde{\delta}^* & -2\tilde{\delta} & \delta\end{pmatrix}. \tag{3.7}$$

The blocks which intermix the three-diagonal representations with one- and two-dimensional representations have the most complicated appearance. They can be written in the form

$$d_{mn}=d_{mn}^{+}-d_{mn}^{-},\quad m=1,4,5,6,\quad n=8,9,$$

$$d_{18}^{\pm}=-d_{49}^{\pm}=i(\varepsilon_{\pm},\lambda^2\tilde{\varepsilon}_{\pm}^*,\lambda\tilde{\varepsilon}_{\pm}),\quad d_{48}^{\pm}=-d_{19}^{\pm}=(\zeta_{\pm},\tilde{\zeta}_{\pm}^*,\tilde{\zeta}_{\pm}),$$

$$d_{58}^{\pm}=-d_{69}^{\pm}=i\begin{pmatrix}\tilde{\varepsilon}_{\pm} & \lambda^{\pm1}\varepsilon_{\pm} & \lambda^2\tilde{\varepsilon}_{\pm}^*\\ \tilde{\varepsilon}_{\pm}^* & \lambda\tilde{\varepsilon}_{\pm} & \lambda^{\mp1}\varepsilon_{\pm}\end{pmatrix},$$

$$d_{68}^{\pm}=-d_{59}^{\pm}=\begin{pmatrix}\tilde{\zeta}_{\pm} & \lambda^{\pm1}\zeta_{\pm} & \lambda^2\tilde{\zeta}_{\pm}^*\\ \tilde{\zeta}_{\pm}^* & \lambda\tilde{\zeta}_{\pm} & \lambda^{\mp1}\zeta_{\pm}\end{pmatrix}. \tag{3.8}$$

The matrix B, which describes the interaction of the a ions with the d ions, has the following form:

$$b_{14}=b_{16}=b_{19}=b_{34}=b_{36}=b_{39}=0,$$

$$b_{11}=\tfrac{1}{2}(\eta_+ +\eta_-)\equiv\eta,\quad b_{15}=\tfrac{1}{2}(\tilde{\eta}_+ +\tilde{\eta}_-,\ \tilde{\eta}_+^* +\tilde{\eta}_-^*), \tag{3.9}$$

$$b_{18}=-\frac{i}{2}(\theta_+ -\theta_-,\tilde{\theta}_+ -\tilde{\theta}_-,\tilde{\theta}_+^* -\tilde{\theta}_-^*).$$

The matrices b_{3m} can be obtained from b_{1m} by changing the signs in front of the terms η_-, $\tilde{\eta}_-$, θ_-, and $\tilde{\theta}_-$. In precisely the same way, the matrices b_{7m} and b_{9m} differ by the sign in front of the combinations having the index "$-$": $\varkappa_-$, $\tilde{\varkappa}_-$, θ_-, etc. We shall therfore give only the blocks of b_{9m}:

$$b_{91}=\frac{1}{2\sqrt{3}}\begin{bmatrix}(\varkappa_+ -\varkappa_-)\\ (\tilde{\varkappa}_+ -\lambda\tilde{\varkappa}_-)^*\\ (\tilde{\varkappa}_+ -\lambda\tilde{\varkappa}_-)\end{bmatrix},$$

$$b_{95}=\frac{1}{2\sqrt{3}}\begin{bmatrix}(\tilde{\varkappa}_+ -\tilde{\varkappa}_-) & (\tilde{\varkappa}_+ -\tilde{\varkappa}_-)^*\\ (\varkappa_+ -\lambda^2\varkappa_-)^* & (\tilde{\varkappa}_+ -\lambda^2\tilde{\varkappa}_-)\\ (\tilde{\varkappa}_+ -\lambda^2\tilde{\varkappa}_-) & (\varkappa_+ -\lambda\varkappa_-)\end{bmatrix},$$

$$b_{98}=\frac{i}{2\sqrt{3}}\begin{bmatrix}(\mu_+ +\mu_-) & (\tilde{\mu}_+ +\tilde{\mu}_-) & (\tilde{\mu}_+ +\tilde{\mu}_-)^*\\ (\tilde{\mu}_+ +\lambda\tilde{\mu}_-)^* & (\mu_+ +\lambda\mu_-) & (\tilde{\mu}_+ +\lambda^2\tilde{\mu}_-)\\ (\tilde{\mu}_+ +\lambda\tilde{\mu}_-) & (\tilde{\mu}_+ +\lambda^2\tilde{\mu}_-)^* & (\mu_+ +\lambda^2\mu_-)\end{bmatrix},$$

$$b_{94}=b_{94}^{(1)}+b_{94}^{(2)},\quad b_{96}=b_{96}^{(1)}+b_{96}^{(2)},\quad b_{99}=b_{99}^{(1)}+b_{99}^{(2)},$$

$$b_{91}^{(1)}=\frac{i}{2\sqrt{3}}\begin{bmatrix}\theta_+ -\theta_-\\ \lambda(\tilde{\theta}_+ -\tilde{\theta}_-)^*\\ \lambda^2(\tilde{\theta}_+ -\tilde{\theta}_-)\end{bmatrix}, \tag{3.10}$$

$$b_{96}^{(1)}=\frac{i}{2\sqrt{3}}\begin{bmatrix}(\tilde{\theta}_+ -\tilde{\theta}_-) & (\tilde{\theta}_+ -\tilde{\theta}_-)^*\\ \lambda(\theta_+ -\theta_-) & \lambda(\tilde{\theta}_+ -\tilde{\theta}_-)\\ \lambda^2(\tilde{\theta}_+ -\tilde{\theta}_-)^* & \lambda^2(\theta_+ -\theta_-)\end{bmatrix},$$

$$b_{91}^{(2)}=-\frac{i}{2\sqrt{3}}\begin{bmatrix}(\mu_+ -\mu_-)\\ \lambda^2(\tilde{\mu}_+ -\tilde{\mu}_-)^*\\ \lambda(\tilde{\mu}_+ -\tilde{\mu}_-)\end{bmatrix},$$

$$b_{96}^{(2)}=-\frac{i}{2\sqrt{3}}\begin{bmatrix}(\tilde{\mu}_+ -\tilde{\mu}_-) & (\tilde{\mu}_+ -\tilde{\mu}_-)^*\\ (\lambda^2\mu_+ -\mu_-) & (\lambda^2\tilde{\mu}_+ -\tilde{\mu}_-)\\ (\lambda^2\tilde{\mu}_+ -\tilde{\mu}_-)^* & (\lambda\mu_+ -\mu_-)\end{bmatrix},$$

$$b_{99}^{(1)}=-\frac{1}{2\sqrt{3}}\begin{bmatrix}(\eta_+ +\eta_-) & (\tilde{\eta}_+ +\tilde{\eta}_-) & (\tilde{\eta}_+ +\tilde{\eta}_-)^*\\ \lambda(\tilde{\eta}_+ +\tilde{\eta}_-)^* & \lambda(\eta_+ +\eta_-) & \lambda(\tilde{\eta}_+ +\tilde{\eta}_-)\\ \lambda^2(\tilde{\eta}_+ +\tilde{\eta}_-) & \lambda^2(\tilde{\eta}_+ +\tilde{\eta}_-)^* & \lambda^2(\eta_+ +\eta_-)\end{bmatrix},$$

$$b_{99}^{(2)}=\frac{1}{2\sqrt{3}}\begin{bmatrix}(\varkappa_+ +\varkappa_-) & (\tilde{\varkappa}_+ +\tilde{\varkappa}_-) & (\tilde{\varkappa}_+ +\tilde{\varkappa}_-)^*\\ (\lambda\tilde{\varkappa}_+ +\varkappa_-)^* & (\lambda^2\varkappa_+ +\varkappa_-) & (\lambda^2\tilde{\varkappa}_+ +\tilde{\varkappa}_-)\\ (\lambda\tilde{\varkappa}_+ +\varkappa_-) & (\lambda^2\tilde{\varkappa}_+ +\varkappa_-)^* & (\lambda\varkappa_+ +\varkappa_-)\end{bmatrix}.$$

One should not be frightened by the cumbersome form of the matrix b_{mn}. At $\mathbf{k}=0$ it simplifies to the utmost, with all of its blocks vanishing except b_{11} and $b_{99}^{(1)}$, which intermix identical representations. The diagonal elements of these blocks will be taken into account exactly. Next in importance are the blocks which are linear in $\mathbf{k}$. These are b_{38}, b_{78}, b_{98}, $b_{74}^{(1)}$, $b_{76}^{(1)}$, $b_{74}^{(2)}$, $b_{76}^{(2)}$, $b_{94}^{(1)}$, and $b_{96}^{(2)}$. The expansion of the matrix elements of the remaining blocks in a series in $\mathbf{k}$ begins at the second order in $\mathbf{k}$ or higher. As a rule, it is sufficient to take them into account in the lowest order of perturbation theory over the entire Brillouin zone.

The A matrix, of dimension 8×8, is constructed rather simply:

$$a_{11}=-a_{33}=2\nu(\mathbf{k}),\quad a_{1m}=0 \text{ for } m\neq1,\quad a_{3m}=0 \text{ for } m\neq3,$$

$$a_{77}=-a_{99}=\begin{bmatrix}-2\rho & \tilde{\rho} & \tilde{\rho}^*\\ \tilde{\rho}^* & \rho & -2\tilde{\rho}\\ \tilde{\rho} & -2\tilde{\rho}^* & \rho\end{bmatrix}. \tag{3.11}$$

Here only a_{11} and a_{33} are large at small k. The expansion of the remaining coefficients begins with terms of at least second order and, as a rule, can be taken into account by perturbation theory.

3.2 *Quasinormal basis.* We recall that in the "irreducible" basis of the Hamiltonian matrix, four pairs of off-diagonal elements remained in the matrix B at $\mathbf{k}=0$. These were $b_{11}(0)=1$ and $b_{9j,9l}=-\delta_{j,l}/3^{1/2}$. These elements can easily be eliminated with the aid of four independent u–v transformations, each of which involves only a single pair of eigenvectors. The coefficients of these transformations can be written in the form

$$u_p(\mathbf{k})=\tfrac{1}{2}[(C_p+|B_p|)^{1/2}+(C_p-|B_p|)^{1/2}](C_p^2-|B_p|^2)^{-1/4},$$

$$v_p(\mathbf{k})=-\frac{B_p}{2|B_p|}[(C_p+|B_p|)^{1/2}-(C_p-|B_p|)^{1/2}](C_p^2-|B_p|^2)^{-1/4}, \tag{3.12}$$

$$C_p=A_p+D_p.$$

Here $B_p\equiv B_{pp}^{(1)}$ is a nonzero element of the matrix $B^{(1)}$ ($p=1$, $p=9.1$; 9.2; 9.3), $A_p\equiv A_{pp}^{(1)}(\mathbf{k})$, and $D_p\equiv D_{pp}^{(1)}(\mathbf{k})$. In the new basis the Hamiltonian matrix is diagonal at $\mathbf{k}=0$ and is nearly diagonal in a significant portion of the Brillouin zone. It is natural to call such a basis "quasinormal." The diagonal elements of the Hamiltonian matrix in this basis give the approximate expressions for the spin-wave frequencies given in formulas (4)–(7) of the Introduction.

§4. PERTURBATION THEORY FOR NONDEGENERATE MODES

4.1 *Ferromagnetic mode.* The dispersion relation of the ferromagnetic mode in the "quasinormal" approximation is given by expressions (6), which were obtained in §2 from simple considerations. This expression gives a good approximation for the frequency $\omega_{d1}^0(\mathbf{k})$ of the ferromagnons over the entire Brillouin zone. Expanding ω_{d1}^0 as $k\to 0$, we find that the coefficient of the $(ak)^2$ term agrees with the exact expression, while the coefficient of $(ak)^4$ is understated by 2%. At the edge of the Brillouin zone the "approximate" formula (6) gives the frequency to within an error of 5%. Of course, in discussing the degree of accuracy we are comparing the approximate expressions not with the true values of the spin-wave frequencies in YIG but with the values that can be obtained by exactly solving the equations of motion (by computer, for instance). With this same accuracy one can expand the trigonometric functions in (2) to order q^4 and obtain a "practical" formula for the frequency from which the small terms have been dropped:

$$\begin{aligned}\omega_{pr}(\mathbf{k}) &= 5|J_{ad}|\{[1+40(1-K/2-2L)q^2 \\ &\quad -(28-2f_4(\mathbf{n}))q^4]^{1/2}-1-2(K-4L)q^2\}, \\ \mathbf{q}&=a\mathbf{k}/8,\quad \mathbf{n}=\mathbf{k}/k,\quad K=J_{dd}/J_{ad},\quad L=J_{aa}/J_{ad}, \\ f_4(\mathbf{n})&=3(n_x^2n_y^2+n_x^2n_z^2+n_y^2n_z^2).\end{aligned} \tag{4.1}$$

In this formula one can readily see the linear part of the k dependence and the low degree of crystallographic anisotropy that appear in the numerical calculation. We note that the linear part of the spectrum is due to the circumstance that YIG is almost an antiferromagnet: For every three d spins there are two a spins with the opposite equilibrium orientation.

In the second order of perturbation theory the correction to the frequency is

$$\omega_{d1}^{(2)}(\mathbf{k})=\omega_{d1}^0(\mathbf{k})-\sum_{m\neq 1,j}\frac{|D_{1,mj}^{(2)}|^2}{\omega_{dmj}^0(\mathbf{k})-\omega_{d1}^0(\mathbf{k})} -\sum_{n\neq 1,l}\frac{|B_{nl,1}^{(2)}|^2}{\omega_{anl}^0(\mathbf{k})+\omega_{d1}^0(\mathbf{k})}. \tag{4.2}$$

Here the matrix elements $D_{1,mj}^{(2)}$ and $B_{nl,1}^{(2)}$ are obtained after a u–v transformation (3.12) of the original matrices (3.3). The first group of terms, proportional to $|D|^2$, arises as a result of the mixing in of the d modes; the second group, proportional to $|B|^2$, from the mixing in of the a modes. All of these terms are nonzero, but the main correction to the frequency ω_{d1}^0 is from the matrix elements proportional to k^2 at small k, namely $D_{1,5j}^{(2)}$ $(j=1,2)$ and $B_{7l,1}^{(2)}$ $(l=1,2,3)$. Taking only these modes into account, we obtain from (4.2)

$$\omega_{d1}^{(2)}(\mathbf{k})=\omega_{d1}^0(\mathbf{k})-\frac{5[J_{dd}u_1(\mathbf{k})-J_{ad}v_1(\mathbf{k})]^2}{\omega_{d5}(0)+\omega_{d1}^0(\mathbf{k})}[1-f_4(\mathbf{n})]q^4 -\frac{40}{27}\frac{J_{ad}^2u_1^2(\mathbf{k})}{\omega_{a7}(0)+\omega_{d1}(\mathbf{k})}f_4(\mathbf{n})q^4. \tag{4.3}$$

The next largest matrix elements are $D_{1,8j}$ $(j=1,2,3)$, whose expansion begins with terms cubic in k. Their contribution to $\omega_{d1}^{(2)}$ is proportional to $(ak)^6$ and falls off rapidly with decreasing k. Furthermore, their contribution goes to zero for $\mathbf{k}\|[111]$ or [100] and, hence, has an additional, angular smallness. Estimates show that this contribution does not exceed 1%, and we shall not even write it out.

4.2 *Antiferromagnetic mode.* The frequency $\omega_{a1}^0(\mathbf{k})$ of the antiferromagnetic mode in the quasinormal approximation is given by the second of expressions (6). The accuracy of this formula is not worse than 15% at the edge of the Brillouin zone. In the second order of perturbation theory

$$\omega_{a1}^{(2)}(\mathbf{k})=\omega_{a1}^0(\mathbf{k})-5\frac{[J_{ad}u_1(\mathbf{k})-J_{dd}v_1(\mathbf{k})]^2}{\omega_{d5}(0)+\omega_{a1}^0(0)}[1-f_4(\mathbf{n})]q^4. \tag{4.4}$$

4.3. The soft d mode ω_{d4} has a minimal gap ~ 100 K for the standard choice of exchange integrals. The formula for the correction to the frequency of this mode is obtained from (4.2) by replacing the index 1 with 4. The largest contribution to the correction is due to the interaction with the ω_{a7} mode:

$$\begin{aligned}\omega_{d4}^{(2)}(\mathbf{k})&=\omega_{d4}^0(\mathbf{k})-\frac{|B_{71,4}|^2+2|B_{72,4}|^2}{\omega_{a7}^0(\mathbf{k})+\omega_{d4}(\mathbf{k})} \\ &=\omega_{d4}^0(\mathbf{k})-10\frac{J_{ad}^2q^2}{\omega_{a7}^0+\omega_{d4}^0(\mathbf{k})}.\end{aligned} \tag{4.5}$$

It is seen that the correction to the frequency amounts to 30% of the dispersion of the mode; allowance for this correction reduces the disagreement between the analytical expression for $\omega_{d4}^{(2)}$ and the numerical results to 10%.

We note that the $\omega_{d4}(\mathbf{k})$ branch crosses the ferromagnetic branch $\omega_{d1}(\mathbf{k})$ at $ak\simeq 2$. The matrix element $|D_{1,4}^{(2)}|$ for the interaction of these oscillations at such values of k is approximately equal to 3 K. The behavior of the functions $\omega_{d1}(\mathbf{k})$ and $\omega_{d4}(\mathbf{k})$ near the crossing is described by the usual formula (see, for example, §39 in Ref. 9). The splitting $2|D_{14}^{(2)}|$ of the spectra is small, and this effect may, as a rule, be neglected.

4.4. The last of the nondegenerate modes, ω_{a3}, has an energy of the order of 1000 K and is never excited at temperatures $T<T_c=560$ K. We shall therefore not write out the correction to the frequency of this mode.

§5. APPROXIMATE CALCULATION OF THE SPECTRA

The procedure described in the previous section for approximate calculations of the spectra is efficient if the off-diagonal matrix elements are small in comparison with the difference (or sum) of the corresponding frequencies. For the majority of matrix elements this condition does in fact hold. However, for frequencies which are close together or degenerate this condition can be violated, and simple perturbation theory is inapplicable. Since the "large" matrix elements are not too great in number, one can use an approximate procedure for diagonalizing the matrices by a sequence of two-dimensional rotations.

To do this we associate with each off-diagonal matrix element through formula (3.12) a coefficient $v_{pp'}$ which characterizes the size of the elementary "rotation" that would cause this element to vanish. We then do the "rotation" corresponding to the maximum $|v_{pp'}|$ and again analyze the resulting Hamiltonian matrix in the same way as before. Then we can again perform the next "largest" rotation. After sev-

eral rotations the Hamiltonian matrix is almost diagonal and then one may use standard perturbation theory. Such a program of analytical calculations will be constructive if the number of large rotations is not too large, so that the expressions obtained for the frequencies are not too unwieldy.

1. We have carried out this program for the symmetric wave-vector directions $\mathbf{k}\|[100]$ and [111]. Here we shall only describe the sequence of two-dimensional rotations, as the reader may find detailed results for the evaluated spectra in our blueprint.[8]

For the direction $\mathbf{k}\|[111]$ the Hamiltonian matrix decomposes into two 4×4 and two 6×6 blocks. One of the small blocks couples the oscillations of the ferromagnetic, antiferromagnetic, and soft d (τ_{d4}) modes and also the a mode τ_{a71}, which is triply degenerate at $\mathbf{k}=0$. This block becomes almost diagonal after a rotation that defines the ferro- and antiferromagnetic oscillations in terms of the vectors τ_{a1} and τ_{d1}. The other small block couples the vectors τ_{a3}, τ_{a91}, τ_{d81}, and τ_{d91}. With the aid of a rotation that intermixes τ_{a91} and τ_{d91} and a second rotation that intermixes τ_{d81} with the new a–d oscillation τ_{d1}, this block is reduced to an almost diagonal form.

The 6×6 blocks are complex conjugates, so it is sufficient to consider one of them, say, that which mixes the vectors τ_{a72}, τ_{a92}, τ_{d51}, τ_{d61}, τ_{d82}, and τ_{d92}. To diagonalize this block one must perform four exact rotations. The first of these intermixes the vectors of identical representations τ_{a92} and τ_{d92} and gives a pair of a–d modes. The next rotation mixes the vectors τ_{d82} and τ_{d92}. Finally, the last two rotations intermix the pairs of vectors τ_{d51} and τ_{d92} and τ_{d61} and τ_{d82}.

After these transformations, all of the frequencies except ω_{d62} are given to within 10% over the entire Brillouin zone. To evaluate ω_{d62} to the same accuracy one must obtain corrections using perturbation theory.

For the direction $\mathbf{k}\|[100]$ it is more convenient to use a different choice of bases for the two-dimensional and three-dimensional representations. After transforming to the new basis the Hamiltonian matrix decomposes into four 3×3 and two 4×4 blocks.

In the first of the 3×3 blocks, which couples vectors τ_{a1}, τ_{d1}, and τ_{d51}, it is sufficient to do a rotation between the vectors τ_{a1} and τ_{d1} of the same representation. As a result, the FM and AFM oscillations will be weakly coupled with τ_{d51}. In the second block, which mixes the vectors τ_{a3}, τ_{d52}, and τ_{d81}, it is necessary to rotate the vectors τ_{d52} and τ_{d81}. The third block couples the vectors τ_{a91}, τ_{d91}, and τ_{d62}. The first rotation is done in the (τ_{a91}, τ_{d91}) plane, and the second rotation in the (τ_{d91}, τ_{d62}) plane.

The last 3×3 block couples the vectors τ_{a71}, τ_{d4}, and τ_{d61}. Here it is sufficient to rotate the vectors τ_{a71} and τ_{d61}.

The first 4×4 block couples the vectors τ_{a73}, τ_{a93}, τ_{d82}, and τ_{d93}. In this block one should perform a rotation in the plane (τ_{a93}, τ_{d93}) of the vectors of the same representation and a rotation of the vectors τ_{d93} and τ_{d82}. The second 4×4 block couples the vectors τ_{a72}, τ_{a92}, τ_{d83}, and τ_{d92} and is the complex conjugate of the first.

2. Of particular interest are the three a–d branches of the d type with activation energy 180 K, which lie next above the soft d mode ω_{d4}. These magnons contribute significantly to the thermodynamics of YIG and to the kinetics of the spin waves of the ferromagnetic branch with $\mathbf{k}\to0$. Unfortunately, one is unable to calculate the spectrum of these three branches for an arbitrary direction of $\mathbf{k}$ with reasonable accuracy and simplicity. The analytical formulas which arise are even more unwieldy than in the case of the symmetric direction [111]. For quantitative treatment we therefore consider only two particular cases. The first—for any value of k but with $\mathbf{k}\|[100]$ or [111]—was considered above. The second—for any direction of $\mathbf{k}$ but with $ak\lesssim1$—we shall consider now. Here we may drop terms higher than second order in k from the expressions for the frequencies. Because of the degeneracy the eigenvectors and $\omega_j(\mathbf{k})$ will not, of course, be analytic functions of $\mathbf{k}$. Detailed calculations of the frequencies $\omega_{d9j}(\mathbf{k})$ are given in our preprint.[8] Here we give only the final expressions for the frequencies:

$$
\begin{aligned}
&\omega_{d9x}=\omega_{d9}(0)+q^2[X-2Zf_4(\mathbf{n})],\\
&\omega_{d9y}=\omega_{d9}(0)+q^2\{Y+Z[f_4(\mathbf{n})-(f_4^2(\mathbf{n})-f_6(\mathbf{n}))^{1/2}]\},\\
&\omega_{d9z}=\omega_{d9}(0)+q^2\{Y+Z[f_4(\mathbf{n})+(f_4^2(\mathbf{n})-f_6(\mathbf{n}))^{1/2}]\},\\
&\omega_{d9}(0)=5|J_{ad}|[(5-2K)^2-8)^{1/2}-1-2K],\\
&X=\xi-2r_6,\quad Y=2\xi-r_8,\quad Z=(\xi+2r_8-3r_6)/3,\\
&\xi=20J_{ad}^2R^{-1},\quad \xi=[20J_{ad}^2-J_{dd}(30J_{ad}-\omega_{d9}(0))]R^{-1},\\
&R=\omega_{d9}(0)/5-J_{ad}-2J_{dd},\\
&r_6=100(uJ_{dd}-2^{1/2}vJ_{ad})^2(10J_{dd}-20J_{ad}-\omega_{d9}(0))^{-1},\\
&r_8=200(vJ_{ad}-2^{1/2}uJ_{dd})^2[20(J_{dd}-J_{ad})-\omega_{d9}(0)]^{-1},\\
&\begin{pmatrix}u\\v\end{pmatrix}=\left(\frac{F\pm1}{2}\right)^{1/2},\quad F=\frac{5-2K}{[(5-2K)^2-8]^{1/2}},\\
&f_6(\mathbf{n})=27n_x^2n_y^2n_z^2.
\end{aligned}
\tag{5.1}
$$

Comparison with the numerical calculation shows that expressions (5.1) have an accuracy not worse than 5% of the dispersion up to $ak\simeq1.2$. This is a small part of the Brillouin zone, but it is important in applications.

§6. THE TEMPERATURE DEPENDENCE OF THE MAGNETIZATION AND THE EXCHANGE INTEGRALS

With the spectrum of the spin waves and the corresponding eigenvectors, one can determine all the thermodynamic properties of the magnetic subsystem of YIG for temperatures not too close to the Curie temperature, i.e., when the interaction between magnons is small. The temperature dependence of the magnetization is the property which has been the most thoroughly studied in experiment,[6] so that a "first-principles" calculation (in the theory of magnetism) of this characteristic is of greater interest.

The magnetization M is proportional to the average values of the density of the z component S_z of the spin:

$$M=2\mu_B\langle S_z\rangle, \tag{6.1}$$

where μ_B is the Bohr magneton, and

$$\langle S_z\rangle=\frac{1}{Nv}\sum_{n}\left\{\sum_{j=9}^{20}(S_0-\langle a_{jn}^{+}a_{jn}\rangle)-\sum_{j=1}^{8}(S_0-\langle a_{jn}^{+}a_{jn}\rangle)\right\}. \tag{6.2}$$

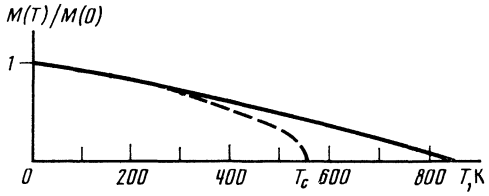

FIG. 4. Temperature dependence of the magnetization for the values of the exchange integrals given in (8). The dashed curve shows the experimental results.

Here v is the volume of the primitive cell, n is the number of the cell, and N is the number of such cells in the crystal. After a Fourier transformation (1.2) we have

$$\langle S_z\rangle = \frac{1}{v}\left\{4S_0 - \sum_{\mathbf{k}}\left[\sum_{j=1}^{8}\langle a_{j\mathbf{k}}^{+}a_{j\mathbf{k}}\rangle - \sum_{j=9}^{20}\langle a_{j\mathbf{k}}^{+}a_{j\mathbf{k}}\rangle\right]\right\}. \quad (6.3)$$

One is readily convinced that the canonical transformations (1.12) leave invariant the form of

$$\sum_{j=1}^{8} a_{j\mathbf{k}}^{+}a_{j\mathbf{k}} - \sum_{j=9}^{20} a_{j\mathbf{k}}^{+}a_{j\mathbf{k}}. \quad (6.4)$$

Therefore

$$\Delta S_z = \langle S_z\rangle_0 - \langle S_z\rangle = \frac{1}{v}\sum_{\mathbf{k}}\left(\sum_{j=9}^{20} n_{j\mathbf{k}} - \sum_{j=1}^{8} n_{j\mathbf{k}}\right), \quad (6.5)$$

where $\langle S_z\rangle_0$ is the value of $\langle S_z\rangle$ at $T=0$, and

$$n_{j\mathbf{k}} \equiv \langle b_{j\mathbf{k}}^{+}b_{j\mathbf{k}}\rangle = [\exp(\omega_j(\mathbf{k})/T) - 1]^{-1}. \quad (6.6)$$

At low temperatures, only the magnons of the lowest, "ferromagnetic" branch are excited. At very low temperatures $T < 25$ K the dispersion relation for these magnons is quadratic, and obviously, $\Delta S_z \propto T^{3/2}$:

$$\Delta S_z = 0.029\,(T/\omega_{ex})^{3/2}. \quad (6.7)$$

For temperatures in the region $25\text{ K} \lesssim T \lesssim 20(|J_{ad}| - 2|J_{dd}|) \simeq 80$ K only magnons of the lowest branch are excited, but most of them are concentrated in the linear region of the dispersion relation, $\omega_{\mathbf{k}} = \omega_l(ak)$, and so $\Delta S_z \propto T^3$, in analogy with the temperature dependence of the number of phonons:

$$\Delta S_z = \frac{\zeta(3)}{2\pi^3}\left(\frac{T}{\omega_l}\right)^3, \quad (6.8)$$

here $\omega_l \simeq 5\sqrt{10}\,|J_{ad}|/4$, in accordance with (4.1).

To determine the temperature dependence of ΔS_z at higher temperatures we used a computer. Starting from the approximate analytical expressions for the frequencies as obtained in the previous sections, the computer performed calculations for any specified set of exchange integrals J_{ad}, J_{dd}, and J_{aa}. Despite the fact that the values of the exchange integrals are poorly known, their combination ω_{ex} in (1) is determined to fair accuracy.[10] It was therefore appropriate to vary only two parameters (J_{ad} and J_{aa}, for example) at fixed ω_{ex}. Changing the parameters in steps $\Delta J_{ad} = \Delta J_{aa} = 0.1$ K, the computer compared the values of the calculated magnetization with the experimental values at $T = 300$ K and $T = 150$ K and, when the difference was less than 1%, put out the values of the exchange integrals. The calculated function $M(T)$ is extremely sensitive to the values of the exchange integrals and at the limits of the interval of admissible values (1.8) had little in common with the experimental curve. It should be pointed out that the magnetization calculated with the values (8) found for the exchange integrals (see Fig. 4) agrees with the experimentally measured value to within 2% at $T = 400$ K, but gives a Curie temperature $T_c = 840$ K that differs from the actual value by 50%. This evidently means that the spin-wave approximation is valid for YIG in the temperature region $0 \leqslant T \lesssim \frac{2}{3} T_c$.

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