Symmetry and magnetic-resonance frequencies in magnetically ordered crystals

V. G. Bar'yakhtar, I. M. Vitebskii, and D. A. Yablonskiĭ

Donetsk Physicotechnical Institute, Ukrainian Academy of Sciences
(Submitted 13 October 1978)
Zh. Eksp. Teor. Fiz. 76, 1381–1391 (April 1979)

A method for the classification and calculation of the frequencies of homogeneous magnetic resonance is developed and makes maximum use of the symmtery of the magnet. The advantages of the new method are particularly pronounced when complex noncollinear magnetic structures are considered. By way of example, all eight resonant frequencies (corresponding to the number of magnetic ions in the unit cell) in a rare-earth orthoferrite are classified by symmetry type for various types of magnetic ordering.

PACS numbers: 76.20. + q

INTRODUCTION

Interest in the study of the high-frequency (HF) properties of magnetically ordered crystals has increased of late. In this connection, new approaches were developed,[1,2] based on general symmetry considerations, using a Lagrangian formalism that makes it possible to obtain all the low-frequency branches of the spectrum. At the same time, the traditional approach based on solving the equations of motion for the magnetic moments of the sublattices (or of the spin operators), using a phenomenological (or microscopical) Hamiltonian, still remains viable. In the calculation of spectra of complex crystals, however, with a large number of magnetic sublattices, the solution of the dispersion equations of high order encounters certain difficulties. It is customary in such cases to combine different magnetic sublattices in one (see, e.g., Ref. 3), but then information on the optical branches of the spectrum is lost.

We demonstrate in this paper a general procedure for calculating the eigenfrequencies of homogeneous oscillations of a spin system, with maximum use made of the magnetic symmetry of the crystal. The equations of motion that determine the HF modes that differ in symmetry are as a rule not intermixed; this lowers substantially, as a rule, the degree of the dispersion equations. In contrast to the generally used approach, the symmetry is taken into account not after the changeover to the small-deviation operators, but in the course of writing down and linearization of the equations of motion. These equations must be written out for irreducible linear combinations of the spin operators, which in general are not small-deviation operators.

The proposed method permits the calculations to be performed for both collinear and noncollinear magnetic structures; in the latter case the advantages of the method become most clearly pronounced. The reason is that in the case of complex noncollinear magnetic structures the transition to the small-deviation operators prior to taking the system symmetry into account is a very cumbersome operation. In our approach, however, we use from the very outset a spin Hamiltonian expressed in terms of irreducible spin operators and having therefore a simplest possible structure. By way of example, we classify by symmetry type and calculate all the eigenfrequencies of the homogeneous magnetic resonance of a rare-earth (RE) orthoferrite for different types of magnetic ordering.

CLASSIFICATION OF THE OSCILLATIONS OF A SPIN SYSTEM

Since small oscillations of a spin system take place against the background of a specified magnetic ordering, they must be also classified in terms of irreducible representations of the magnetic symmetry group of the crystal (as is done, for example, when molecule vibrations are considered[4]). The number of resonant modes of a given symmetry is not directly connected with the magnetic symmetry of the crystal. To determine this number one must invoke information on the concrete magnetic structure (know the positions of the magnetic ions), and resort also to model considerations. In the present paper we treat the spin-system oscillations in the spin-wave approximation on the basis of a quadratic spin Hamiltonian.

To find the resonant modes of a system consisting of n magnetic sublattices we can start from the equations of motion for the spin operators of the individual sublattices, wherein $3n$ equations for the $3n$ components of the spin operators determine n resonant modes (just as the three equations for $\hat{S}_x$, $\hat{S}_y$, and $\hat{S}_z$ in a ferromagnet determine a single ferromagnetic-resonance frequency). To prevent intermixing of the equations that describe oscillations of different symmetry, we write them down not for the components of the sublattice spin operators, but for $3n$ of their linear combinations, which realize irreducible representations of the magnetic symmetry group of the crystal (irreducible operators).

The number k_i of resonant modes that transform in accord with the i-th irreducible representations is usually equal to $k_i = n_i/3$, where n_i is the number of irreducible operators that transform in accord with the i-th representation. However, in the case when the spin of certain sublattices in the equilibrium state are oriented along symmetrical crystallographic directions, the equality $k_i = n_i/3$ may possibly not hold. The reason is that in the spin-wave approximation there are now longitudinal oscillations of the sublattice magnetization. If l_i is the dimensionality of the i-th representation, then the number of different frequencies is equal to k_i/l_i, inasmuch as each frequency l_i is multiply degenerate. In the next section we shall illustrate all the

 0038-5646/79/040703-06$02.40

foregoing in detail, using as an example RE orthoferrites.

FREQUENCIES OF HOMOGENEOUS RESONANCE OF RE ORTHOFERRITIES

RE orthoferrites (chemical formula $RFeO_3$, where R stands for an RE ion and Fe can be replaced by Cr or Co) belong to the space group D_{2h}^{16}. The unit cell contains four $RFeO_3$ molecules. Below $T_N \sim 600$ K the iron subsystem has in all cases antiferromagnetic ordering (AF) of G type (see Table I) with a weak admixture of other modes (the possible types of magnetic symmetry of RE orthoferrites were investigated in Ref. 3). Using these data and the scheme described in the preceding section, we present a classification of the homogeneous oscillations of the spin system by spin types.

In column 2 of Table II is indicated the magnetic symmetry of the oscillating part of the magnetic sublattices; it corresponds to one of the irreducible representations of the group of magnetic symmetry of the crystal (these representations can be found in standard fashion on the basis of Table I). Column 3 of Table II indicates the number of resonant modes of corresponding symmetry (since the group is Abelian, all frequencies are nondegenerate).

In column 4 of Table II is given an arbitrary classification of the resonant frequencies for the case when the principal interaction is an isotropic exchange interaction between the Fe ions, which leads to AF ordering of G type in the iron system (the Γ_3 phase is not realized in this case). This condition is satisfied for all orthoferrites without exception. We shall arbitrarily call modes optical (O) or acoustic (A) if they are connected with the Fe sublattices, and designate by EPR the modes connected with the RE subsystem.[1)] This distinction is meaningful so long as the corresponding frequencies differ substantially. The optical modes of antiferromagnetic resonance (AFMR) lie much higher than the remaining one, a fact that we shall make use of in the calculations. The acoustic frequencies, in turn, usually

TABLE I. Magnetic configurations* that are reducible with respect to the crystallographic group D_{2h}^{16}.

	Fe			R				1	2_{1x}	2_{1y}	2_{1z}	$\bar{1}$	$\bar{2}_{1x}$	$\bar{2}_{1y}$	$\bar{2}_{1z}$
Γ_1	A_x	G_y	C_z				C_{Rz}	+	+	+	+	+	+	+	+
Γ_2	F_x	C_y	G_z	F_{Rx}	C_{Ry}			+	+	−	−	+	+	−	−
Γ_3	C_x	F_y	A_z	C_{Rx}	F_{Ry}			+	−	+	−	+	−	+	−
Γ_4	G_x	A_y	F_z				F_{Rz}	+	−	−	+	+	−	−	+
Γ_5				G_{Rx}	A_{Ry}			+	+	+	+	−	−	−	−
Γ_6							A_{Rz}	+	+	−	−	−	−	+	+
Γ_7							G_{Rz}	+	−	+	−	−	+	−	+
Γ_8				A_{Rx}	G_{Ry}			+	−	−	+	−	+	+	−

*We use here

$F = S_1 + S_2 + S_3 + S_4$, $F_R = s_5 + s_6 + s_7 + s_8$,
$C = S_1 + S_2 - S_3 - S_4$, $C_R = s_5 + s_6 - s_7 - s_8$,
$G = S_1 - S_2 + S_3 - S_4$, $G_R = s_5 - s_6 + s_7 - s_8$,
$A = S_1 - S_2 - S_3 + S_4$, $A_R = s_5 - s_6 - s_7 + s_8$.

In addition to the indicated pure (irreducible) configurations, mixed (reducible) configurations with several indices are possible, indicating irreducible components (e.g., Γ_{12}).

TABLE II. Classification of the frequencies of homogenous magnetic resonance in $RFeO_3$ by symmetry types

Symmetry of magnetic ordering	Symmery of homogeneous oscillations	Number of resonant modes	AF ordering of type G*	$\chi_{\alpha\beta}$ components having a pole
Γ_1	Γ_1	1	O	–
	Γ_2	2	A+EPR	χ_{xx}
	Γ_3	2	O+EPR	χ_{yy}
	Γ_4	1	A	χ_{zz}
	Γ_5	1	EPR	–
	Γ_6	0	–	–
	Γ_7	0	–	–
	Γ_8	1	EPR	–
Γ_2	Γ_{12}	3	O+A+EPR	χ_{xx}
	Γ_{34}	3	O+A+EPR	χ_{yy}, χ_{zz}, χ_{yz}
	Γ_{56}	1	EPR	–
	Γ_{78}	1	EPR	–
Γ_3	Γ_{13}	3	Not realized	χ_{yy}
	Γ_{24}	3		χ_{xx}, χ_{zz}, χ_{xz}
	Γ_{57}	1		–
	Γ_{68}	1		–
Γ_4	Γ_{14}	2	O+A	χ_{zz}
	Γ_{23}	4	O+A+2EPR	χ_{xx}, χ_{yy}, χ_{xy}
	Γ_{58}	2	2EPR	–
	Γ_{67}	0	–	–

*For the notation see the text.

exceed the EPR frequencies (in the case of Kramers RE ions), but this fact is not used in the calculations. Finally, the last column of Table II indicates the HF magnetic-susceptibility tensor components $\chi_{\alpha\beta}$ whose poles correspond to the oscillations of the given symmetry. A dash in this column means that the indicated oscillations are not connected with the oscillations of the resultant magnetic moment of the sample, and consequently can not be excited by a homogeneous high-frequency field. The reason is that none of the components of the magnetic field H are transformed in accord with the corresponding irreducible representations.

We proceed now to calculate the resonant frequencies. We write down the Hamiltonian of the system in the form

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_{Fe-Fe} + \hat{\mathcal{H}}_{Fe-R} + \hat{\mathcal{H}}_{R-R} = \sum K^{\alpha\beta}_{m\mu,n\nu} \hat{S}^{\alpha}_{m\mu} \hat{S}^{\beta}_{n\nu} + \sum L^{\alpha\beta}_{m\mu,n\xi} \hat{S}^{\alpha}_{m\mu} \hat{s}^{\beta}_{n\xi} + \sum \Lambda^{\alpha\beta}_{m\xi,n\zeta} \hat{s}^{\alpha}_{m\xi} \hat{s}^{\beta}_{n\zeta}, \quad (1)$$

where the spin operators $\hat{S}^{\alpha}_{m\mu}$ and $\hat{s}^{\alpha}_{m\xi}$ pertain respectively to the ions Fe and R; $\alpha, \beta = x, y, z$; m and n are the numbers of the unit cells; $\mu, \nu = 1, 2, 3, 4$ and $\xi, \zeta = 5, 6, 7, 8$ are respectively the numbers of the positions of the Fe and R ions in the unit cell. In the case of the RE ions the effect of the crystal field causes the dimensionality of the "effective" spin at low temperatures to be 1/2 (Ref. 6) (this pertains mainly to Kramers ions). We shall therefore take $\hat{\mathbf{s}}_{m\xi}$ to mean precisely the effective-spin operator. The operator of the magnetic moment of the RE ion is connected with the operator $\hat{\mathbf{s}}_{m\xi}$ via the anisotropic g factor; this circumstance must be borne in mind when the susceptibility is calculated.

Since we are interested only in homogeneous states (i.e., $\mathbf{k} = 0$) of the spin system at $T = 0$, we can consider in place of (1) the Hamiltonian

$$\hat{\mathcal{H}}^{(0)} = \sum K^{\alpha\beta}_{\mu\nu} \hat{S}^{\alpha}_{\mu} \hat{S}^{\beta}_{\nu} + \sum L^{\alpha\beta}_{\mu\xi} \hat{S}^{\alpha}_{\mu} \hat{s}^{\beta}_{\xi} + \sum \Lambda^{\alpha\beta}_{\xi\zeta} \hat{s}^{\alpha}_{\xi} \hat{s}^{\beta}_{\zeta}, \quad (2)$$

where

$$\hat{S}^{\alpha}_{\mu} = \sum_m \hat{S}^{\alpha}_{m\mu}, \quad \hat{s}^{\alpha}_{\xi} = \sum_m \hat{s}^{\alpha}_{m\xi};$$

$$K^{\alpha\beta}_{\mu\nu} = \frac{1}{N} \sum_m K^{\alpha\beta}_{m\mu,n\nu}, \quad L^{\alpha\beta}_{\mu\xi} = \frac{1}{N} \sum_m L^{\alpha\beta}_{m\mu,n\xi}, \quad \Lambda^{\alpha\beta}_{\xi\zeta} = \frac{1}{N} \sum_m \Lambda^{\alpha\beta}_{m\xi,n\zeta}.$$

Each of the coefficients K, L, and Λ in (2) takes on 144 values (when account is taken of the numbers of the sublattices and of the coordinate indices). Because of the lattice symmetry, some of them are equal to one another. Using these relations, which are determined completely by the symmetry elements of the D_{2h}^{16} group and by the positions of the Fe and R ions in the unit cell, we transform the Hamiltonian (2) into

$$\hat{\mathcal{H}}^{(0)}=\hat{\mathcal{H}}^{(0)}_{Fe-Fe}+\hat{\mathcal{H}}^{(0)}_{Fe-R}+\hat{\mathcal{H}}^{(0)}_{R-R}, \tag{3}$$

where

$$\hat{\mathcal{H}}^{(0)}_{Fe-Fe}=I_{1x}\hat{A}_x^2+I_{1y}G_y^2+I_{1z}\hat{C}_z^2+D_{1z}\hat{A}_x\hat{G}_y+D_{1y}\hat{A}_x\hat{C}_z+D_{1z}\hat{G}_y\hat{C}_z+I_{1z}\hat{F}_x^2$$
$$+I_{2y}\hat{C}_y^2+I_{3z}\hat{G}_z^2+D_{2z}\hat{F}_x\hat{G}_y+D_{2y}\hat{F}_x\hat{C}_z+D_{2z}\hat{C}_y\hat{G}_z+I_{2x}\hat{G}_x^2+I_{1y}\hat{F}_y^2+I_{4z}\hat{A}_z^2$$
$$+D_{3z}\hat{C}_x\hat{F}_y+D_{3y}\hat{C}_x\hat{A}_z+D_{3x}\hat{F}_y\hat{A}_z+I_{3x}\hat{G}_x^2+I_{4y}\hat{A}_y^2+I_{1z}\hat{F}_z^2+D_{4z}\hat{G}_x\hat{A}_y$$
$$+D_{4y}\hat{G}_x\hat{F}_z+D_{4x}\hat{A}_y\hat{F}_z, \tag{4}$$

$$\hat{\mathcal{H}}^{(0)}_{Fe-R}=Q_{xz}\hat{A}_x\hat{C}_{Rz}+Q_{yz}\hat{G}_y\hat{C}_{Rz}+Q_{zz}\hat{C}_z\hat{C}_{Rz}+P_{xx}\hat{F}_x\hat{F}_{Rx}+P_{yx}\hat{C}_y\hat{F}_{Rx}$$
$$+P_{zx}\hat{G}_z\hat{F}_{Rx}+Q_{zy}\hat{F}_z\hat{C}_{Ry}+Q_{yy}\hat{C}_y\hat{C}_{Ry}+Q_{zy}\hat{G}_z\hat{C}_{Ry}+Q_{zx}\hat{C}_z\hat{C}_{Rx}$$
$$+Q_{yx}\hat{F}_y\hat{C}_{Rz}+Q_{zx}\hat{A}_z\hat{C}_{Rx}+P_{xy}\hat{C}_x\hat{F}_{Ry}+P_{yy}\hat{F}_y\hat{F}_{Ry}+P_{zy}\hat{A}_z\hat{F}_{Ry}+P_{zx}\hat{G}_x\hat{F}_{Rz}$$
$$+P_{yz}\hat{A}_y\hat{F}_{Rz}+P_{zz}\hat{F}_z\hat{F}_{Rz}, \tag{5}$$

$$\hat{\mathcal{H}}^{(0)}_{R-R}=V_{zz}{}^R\hat{C}_{Rz}^2+U_{xx}{}^R\hat{F}_{Rx}^2+V_{yy}{}^R\hat{C}_{Ry}^2+V_{xy}{}^R\hat{F}_{Rx}\hat{C}_{Ry}+U_{yy}{}^R\hat{F}_{Ry}^2$$
$$+V_{xx}{}^R\hat{C}_{Rx}^2+U_{xy}{}^R\hat{C}_{Rx}\hat{F}_{Ry}+U_{zz}{}^R\hat{F}_{Rz}^2+W_{xx}{}^R\hat{G}_{Rx}^2+Y_{yy}{}^R\hat{A}_{Ry}^2+Y_{xy}{}^R\hat{G}_{Rx}\hat{A}_{Ry}$$
$$+Y_{zz}{}^R\hat{A}_{Rz}^2+Y_{xx}{}^R\hat{A}_{Rx}^2+W_{yy}{}^R\hat{G}_{Ry}^2+W_{xy}{}^R\hat{A}_{Rx}\hat{G}_{Ry}+W_{zz}{}^R\hat{G}_{Rz}^2. \tag{6}$$

The operators $\hat{F}_\alpha$, $\hat{C}_\alpha$, $\hat{G}_\alpha$, $\hat{A}_\alpha$, $\hat{F}_{R\alpha}$, $\hat{C}_{R\alpha}$, $\hat{G}_{R\alpha}$, and $\hat{A}_{R\alpha}$ in (4)–(6) are expressed in terms of $\hat{S}_\mu^\alpha$ and $\hat{\mathfrak{S}}_\xi^\alpha$ in exactly the same manner as their mean values (see Table I); to distinguish them from the latter, all the operator quantities are marked here and below by a caret. The magnetic-interaction constants in (4)–(6) are linearly expressed in terms of the constants, K, L, and Λ of the Hamiltonian (2). Taking Table I into account, it is easily noted that all the terms in (4)–(6) are invariant to transformations of the D_{2h}^{16} group. Thus, the new constants I, D, P, Q, U^R, V^R, W^R, and Y^R are already independent (their total number is 58 as against the 3×144 constants, K, L, and Λ).

We note finally that the parameters I in (4) include the isotropic exchange interaction between the Fe ions, so that the following inequality is valid:

$$|I_{1\alpha}|\gg I_{1\alpha}-I_{1\beta},\ D,\ P,\ Q,\ U^R,\ V^R,\ W^R,\ Y^R, \tag{7}$$

and similarly for $I_{2\alpha}$, $I_{3\alpha}$, and $I_{4\alpha}$. In addition,

$$|I_{3\alpha}|=-I_{3\alpha}>-I_{1\alpha},\ -I_{2\alpha},\ -I_{4\alpha} \tag{8}$$

which is the condition for the realization of AF ordering of type G.

We proceed to calculate the resonant frequencies. To prevent intermixing of HF modes of different symmetry, we shall write the equations of motion not for the spin operators of the individual sublattices, but for their irreducible combinations that were introduced above. Since the group D_{2h}^{16} is Abelian, these operators are irreducible also with respect to any of the possible magnetic-symmetry groups of the ground state of the orthoferrites.

The equations of motion take the standard form[2)]

$$i\dot{\hat{a}}=[\hat{a},\ \hat{\mathcal{H}}], \tag{9}$$

where $\hat{a}$ is one of the irreducible operators introduced above. As a result of commutation, terms that are quadratic in the irreducible operators appear in the right hand side of (9). We shall linearize Eq. (9) in the spirit of the random-phase approximation, i.e., we shall replace the product of two operators $\hat{a}\hat{b}$ in accord with the scheme

$$\hat{a}\hat{b}\to\langle\hat{a}\rangle\langle\hat{b}\rangle+\langle\hat{a}\rangle\hat{b}+\langle\hat{b}\rangle\hat{a}. \tag{10}$$

This procedure corresponds at $T=0$ to the spin-wave approximation. (The terms of the type $\langle\hat{a}\rangle\langle\hat{b}\rangle$ cancel out in the equations of motion of the ground state is suitably chosen.) We note that the substitution (10) is fully analogous to the classical linearization procedure, when the product of two quantities ab is replaced by the expression

$$ab=(a_0+\tilde{a})(b_0+\tilde{b})\to a_0b_0+a_0\tilde{b}+b_0\tilde{a}.$$

The irreducible operators satisfy the following simple commutation relations:

$$[\hat{F}_x\hat{F}_y]=[\hat{A}_x\hat{A}_y]=[\hat{G}_x\hat{G}_y]=[\hat{C}_x\hat{C}_y]=iF_z,\quad [\hat{F}_x\hat{A}_y]=[\hat{C}_x\hat{G}_y]=i\hat{A}_z,$$
$$[\hat{F}_x\hat{C}_y]=[\hat{G}_x\hat{A}_y]=i\hat{C}_z,\quad [\hat{F}_x\hat{G}_y]=[\hat{C}_x\hat{A}_y]=i\hat{G}_z, \tag{11}$$

and the remaining relations are obtained by cyclic permutation of the indices.

Taking (4)–(6) and (11) into account we can write down the equations of motion for all 24 irreducible operators introduced above. The result of the distribution (10) is determined by the magnetic symmetry of the crystal, i.e., it depends on the concrete method of magnetic ordering. For example, in the phase Γ_1 only the mean values of the operators $\hat{A}_x$, $\hat{G}_y$, $\hat{C}_z$, and $\hat{C}_{Rz}$ differ from zero (see Table I), and the mean values of the operators $\hat{F}_x$, $\hat{C}_y$, $\hat{G}_z$, $\hat{C}_{Ry}$, and $\hat{F}_{Rx}$ appear in the phase Γ_{12}. The structure of this system reflects not only the lattice symmetry, but also the symmetry of the magnetic ordering. For example, if the symmetry of the phase in which the oscillations are considered is Γ_2, then the system of equations breaks up automatically into four independent blocks (in accord with the four irreducible representations of the magnetic-symmetry group Γ_2, see Table II). Analogously, in the case of the phase Γ_1 the number of independent blocks is equal to eight, and in the phase Γ_{24} it is equal only to two. We note that wherever we refer to a magnetic symmetry group we have in mind only the spatial symmetry elements, (i.e., all or some of the elements indicated in Table I). On the other hand, the introduction of the time-reversal operation $\hat{R}$ yields in this case no additional information whatever concerning the resonant modes.

If we now replace in the obtained system of 24 equations the mean values of the irreducible operators by their equilibrium values obtained from the condition of the minimum of the energy of the ground state, then this system is left with only 16 independent equations. The corresponding secular equation for the determination of the resonant frequencies is an equation of eight degree in E^2 and determines eight resonant modes (in accord with the number of magnetic sublattices).

Investigating separately each block of coupled equations, we obtain all the frequencies of the oscillations of

the corresponding symmetry. We now demonstrate the procedure described above with a concrete example. Let the magnetic ordering symmetry be Γ_4. We consider first the symmetric oscillations of the spin system, i.e., the oscillations with symmetry Γ_{14}. To this end we write down the equations of motion for the operator $\hat{G}_x$, since it transforms in accord with the representation Γ_4. After the uncoupling (with account taken of the fact that only the mean values G_x, A_y, F_z, and F_{Rx} differ from zero), we find that $\hat{A}_x$, $\hat{G}_y$, and $\hat{C}_z$ also become intermixed in the equation for $\hat{G}_x$; we write down the equation of motion for each of them. As a result, after decoupling, we obtain a closed system of six linear equations for the operators $\hat{G}_x$, $\hat{A}_y$, $\hat{F}_z$, $\hat{A}_x$, $\hat{G}_y$, and $\hat{C}_z$. We note that this system does not include the operators $\hat{F}_{Rz}$ and $\hat{C}_{Rz}$, although they should enter in this block from symmetry considerations. This is caused by the use of the spin-wave approximation (the absence of longitudinal oscillations of the sublattice magnetization). Thus, the RE subsystem does not participate in oscillations of symmetric type in the Γ_4 phase.

Equating to zero the determinant of the system, we obtain a fourth-degree equation in E^2. If the mean values G_x, A_y, and F_z are replaced by their equilibrium values, then the free term in the secular equation vanishes and we obtain an equation quadratic in E^2. To write down the answers we shall use the following obvious approximations: 1) the isotropic exchange between the Fe ions greatly exceeds all other interactions [inequality (7)]; 2) the noncollinearity of four iron sublattices is due to antisymmetric exchange interaction between the iron ions, which is much stronger than the single-ion anisotropy and the dipole interaction (this is always true for $3d$ ions with singlet orbital state). We make no simplifying assumptions whatever with respect to the interactions $\mathcal{H}_{Fe\text{-}R}$ and $\mathcal{H}_{R\text{-}R}$. We emphasize that these approximations serve only one purpose—to simplify the determination of the equilibrium directions of the sublattice magnetizations in the iron subsystem.

In the phase Γ_4 at $T=0$ we then obtain

$$G_x \approx 4S, \quad A_y \approx -D_{xz}G_x/2(I_1-I_3), \quad F_z \approx -D_{xy}G_x/2(I_1-I_3), \quad F_{Rz}=4s. \tag{12}$$

The frequencies of the symmetric oscillations in the phase Γ_4 are

$$E_{14}^{(4)}(1)=2G_x[(I_{2z}-I_{3x})(I_{4y}-I_{3x})]^{1/2}, \tag{13}$$

$$E_{14}^{(4)}(2)=G_x\left[2(I_1-I_3)\left[2(I_{3y}-I_{3x})+\frac{D_{4y}^2}{2(I_1-I_3)}-\frac{D_{1x}^2}{2(I_2-I_3)}-P_{xz}F_{Rz}\right]\right]^{1/2}. \tag{14}$$

The HF mode $E_{14}^{(4)}(1)$ is optical, while $E_{14}^{(4)}(2)$ is acoustic; G_x and F_{Rz} are given in (12). In accord with the inequality (7), the coordinate index of the constant I can sometimes be left out. The terms with the parameter D in (14) determine the renormalization of the acoustic mode because of the mixing of the configurations of type F, C, and A, while the term $P_{xz}F_{Rz}$ describes the influence of the RE of the subsystem (although this subsystem is not involved in this case in the oscillatory process itself).

We consider now oscillations with symmetry Γ_{23} (the magnetic-ordering symmetry is as before Γ_4). The symmetry Γ_{23} corresponds to four resonant modes (see Table II), so that in the general case it is necessary to solve a fourth-degree secular equation. We confine ourselves, however, for simplicity, to the case of a nonmagnetic RE ion (e.g., $YFeO_3$ or $LaFeO_3$). The system of equations describing this oscillation mode then contains the operators $\hat{F}_x$, $\hat{C}_y$, $\hat{G}_z$, $\hat{C}_x$, $\hat{F}_y$, and $\hat{A}_z$. Following exactly the preceding procedure, we obtain for the frequencies (without the contribution of the RE subsystem)

$$E_{23}^{(4)}(3)=2G_x[(I_{2y}-I_{3x})(I_{4z}-I_{3x})]^{1/2}, \tag{15}$$

$$E_{23}^{(4)}(4)=G_x\left[2(I_1-I_3)\left[2(I_{3z}-I_{3x})+\frac{D_{4z}^2}{2(I_1-I_3)}-\frac{D_{1z}^2}{2(I_2-I_3)}\right]\right]^{1/2}, \tag{16}$$

The first of the frequencies is optical and the second is acoustic. We note that in the exchange approximation, i.e., in the absence of anisotropy) the two optical modes with different symmetry have the same frequency, but the latter is, of course, the same in the different magnetic configurations.

The remaining two HF modes have the symmetry Γ_{58} and describe the oscillations of the spin system with violation of the inversion center $\bar{1}$. Since the positions of the Fe ions coincide with the crystal-lattice inversion centers, only the RE subsystem take part in these oscillations. The closed system of linear equations includes the operators $\hat{G}_{Rx}$, $\hat{A}_{Ry}$, $\hat{A}_{Rx}$, and $\hat{G}_{Ry}$. Since we make no simplifying assumptions whatever concerning the interaction constants in $\mathcal{H}_{Fe\text{-}R}$ and $\mathcal{H}_{R\text{-}R}$, the results obtained for the frequencies are rather unwieldy (although their derivation is straightforward).

We thus have for the frequencies of the oscillations of type Γ_{58} in the Γ_4 phase

$$E_{58}^{(4)}(7,8)=\left[\gamma\pm\left\{\gamma^2-\left([2(W_{yy}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x][2(Y_{xx}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x]-(W_{xy}^R)^2F_{Rz}^2\right)\left([2(Y_{yy}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x][2(W_{xx}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x]-(Y_{xy}^R)^2F_{Rz}^2\right)\right\}^{1/2}\right]^{1/2}, \tag{17}$$

where

$$2\gamma=[2(Y_{yy}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x][2(Y_{xx}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x]+[2(W_{xx}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x][2(W_{yy}^R-U_{zz}^R)F_{Rz}-P_{xz}G_x]-2Y_{xy}^RW_{xy}^RF_{Rz}^2.$$

If we neglect the $\mathcal{H}_{R\text{-}R}$ interaction, this expression becomes much simpler:

$$E_{58}^{(4)}(7.8)=|P_{xz}G_x|,$$

i.e., these frequencies are degenerate in this approximation.

It is left for us to consider the configurations Γ_1 and Γ_2. The procedure for calculating the homogeneous-resonance frequencies was described in detail above. Since the equations of motion are written for irreducible operators, they break up automatically after linearlization into independent groups that describe the oscillations of a spin system of definite symmetry. The possibly symmetry types of these oscillations can be identified immediately by determining the irreducible representations of the magnetic-symmetry group of a particular magnetic phase. We present below the results for

the resonant frequencies in the phases Γ_1 and Γ_2. The upper and lower indices label respectively the symmetry of the magnetic ordering and the symmetry of the HF mode (i.e., the symmetry of the oscillating part of the sublattice moments). The quantity in the parentheses is the arbitrary number of the resonant frequency.

The phase Γ_1:

$$G_y \approx 4S, \quad A_x \approx -D_{1x}G_y/2(I_4-I_3), \quad C_z \approx -D_{1x}G_y/2(I_2-I_3), \quad C_{Rz}=4s; \tag{18}$$

$$E_1^{(1)}(1)=2G_y[(I_{2z}-I_{3y})(I_{4x}-I_{3y})]^{1/2}; \tag{19}$$

$$E_2^{(1)}(2,3)=[1/2(\omega_1^2+\Omega_1^2)\pm\{1/4(\omega_1^2-\Omega_1^2)^2+Q_{yz}(Q_{zy}^2+P_{zx}^2)G_y^2C_{Rz} +2[Q_{zy}^2(V_{zz}^R-U_{xx}^R)+P_{zx}^2(V_{zz}^R-V_{yy}^R)+Q_{zy}P_{zx}V_{xy}^R]G_yC_{Rz}^2\}^{1/2}]^{1/2}, \tag{20}$$

where

$$\Omega_1^2=2G_y^2(I_1-I_3)\left[2(I_{3z}-I_{3y})+\frac{D_{1z}^2}{2(I_4-I_3)}-\frac{D_{2y}^2}{2(I_1-I_3)}-Q_{yz}\frac{C_{Rz}}{G_y}\right],$$

$$\omega_1^2=[2(U_{xx}^R-V_{zz}^R)C_{Rz}-Q_{yz}G_y][2(V_{yy}^R-V_{zz}^R)C_{Rz}-Q_{yz}G_y]-(V_{xy}^R)^2C_{Rz}^2;$$

$$E_3^{(1)}(4)=2G_y[(I_{2x}-I_{3y})(I_{4x}-I_{3y})]^{1/2}; \tag{21}$$

$$E_3^{(1)}(5)=[[2(U_{yy}^R-V_{zz}^R)C_{Rz}-Q_{yz}G_y][2(V_{xx}^R-V_{zz}^R)C_{Rz} -Q_{yz}G_y]-(U_{xy}^R)^2C_{Rz}^2]^{1/2}; \tag{22}$$

$$E_4^{(1)}(6)=G_y\left[2(I_1-I_3)\left[2(I_{3x}-I_{3y})+\frac{D_{1y}^2}{2(I_2-I_3)}-\frac{D_{2y}^2}{2(I_1-I_3)}-Q_{yz}\frac{C_{Rz}}{G_y}\right]\right]^{1/2}; \tag{23}$$

$$E_5^{(1)}(7)=[[2(Y_{yy}^R-V_{zz}^R)C_{Rz}-Q_{yz}G_y][2(W_{xx}^R-V_{zz}^R)C_{Rz} -Q_{yz}G_y]-(Y_{xy}^R)^2C_{Rz}^2]^{1/2}; \tag{24}$$

$$E_6^{(1)}(8)=[[2(W_{yy}^R-V_{zz}^R)C_{Rz}-Q_{yz}G_y][2(Y_{xx}^R-V_{zz}^R)C_{Rz} -Q_{yz}G_y]-(W_{xy}^R)^2C_{Rz}^2]^{1/2}. \tag{25}$$

The principal information on these frequencies is contained in Table II. We add only that the quantity $Q_{yz}G_y$, which is frequently encountered in these equations, has the meaning of the effective field produced by the iron at the RE ions, and $Q_{yz}G_{Rz}$ is conversely the effective field of the RE ions at the Fe ions.

The dynamic coupling of the RE subsystem with the iron subsystem occurs for oscillations with symmetry Γ_2 [see (20)].

The phase Γ_2:

$$G_z \approx 4S, \quad F_x \approx -D_{2y}G_z/2(I_1-I_3), \quad C_y \approx -D_{2x}G_z/2(I_2-I_3),$$
$$G_z(Q_{zy}F_{Rx}-P_{zx}C_{Ry})+2(V_{yy}^R-U_{xx}^R)F_{Rx}C_{Ry}+V_{xy}^R(F_{Rx}^2-C_{Ry}^2)=0, \tag{26}$$
$$F_{Rx}^2+C_{Ry}^2=(4s)^2,$$

the last expression in (26) determines the equilibrium directions of the RE sublattices in the $x-y$ plane;

$$E_{12}^{(2)}(1)=2G_z[(I_{2y}-I_{3z})(I_{4x}-I_{3z})]^{1/2}; \tag{27}$$

$$E_{12}^{(2)}(2,3)=[1/2(\omega_2^2+\Omega_2^2)\pm\{1/4(\omega_2^2-\Omega_2^2)^2+2G_z^2Q_{yz}^2(I_1-I_3)[2(U_{xx}^R -V_{yy}^R)(C_{Ry}^2-F_{Rx}^2)-4V_{xy}^RF_{Rx}C_{Ry}-G_z(P_{zx}F_{Rx}+Q_{zy}C_{Ry})]\}^{1/2}]^{1/2}, \tag{28}$$

where

$$\Omega_2^2=2G_z^2(I_4-I_3)[2(I_{3y}-I_{3z})+D_{2y}^2/2(I_1-I_3)-D_{1z}^2/2(I_4-I_3)-P_{zx}F_{Rx}-Q_{zy}C_{Ry}]$$
$$\omega_2^2=[2(U_{xx}^R-V_{yy}^R)C_{Ry}-2V_{xy}^RF_{Rx}-Q_{zy}G_z][2(V_{zz}^R-V_{yy}^R)C_{Ry} -V_{xy}^RF_{Rx}-Q_{zy}G_z]+[2(V_{zz}^R-U_{xx}^R)F_{Rx}-V_{xy}^RC_{Ry}-P_{zx}G_z][2(V_{yy}^R -U_{xx}^R)F_{Rx}-2V_{xy}^RC_{Ry}-P_{zx}G_z];$$
$$E_{23}^{(3)}(4)=2G_z[(I_{2x}-I_{3z})(I_{4y}-I_{3z})]^{1/2}; \tag{29}$$

$$E_{34}^{(3)}(5,6)=[1/2(\omega_3^2+\Omega_3^2)\pm\{1/4(\omega_3^2-\Omega_3^2)^2+2G_z^2P_{xx}^2(I_1-I_3)[2(V_{xx}^R -V_{yy}^R)C_{Ry}^2-2(U_{xx}^R-U_{yy}^R)F_{Rx}^2-2(V_{xy}^R+U_{xy}^R)F_{Rx}C_{Ry} -G_z(P_{zx}F_{Rx}+Q_{zy}C_{Ry})]\}^{1/2}]^{1/2}, \tag{30}$$

where

$$\Omega_3^2=2G_z^2(I_1-I_3)[2(I_{3x}-I_{3z})+D_{2x}^2/2(I_2-I_3)-D_{1z}^2/2(I_4-I_3)-Q_{zy}C_{Ry}-P_{zx}F_{Rx}].$$
$$\omega_3^2=[2(U_{yy}^R-U_{xx}^R)F_{Rx}-(V_{xy}^R+U_{xy}^R)C_{Ry}-P_{zx}G_z][2(U_{zz}^R-U_{xx}^R)F_{Rx} -V_{xy}^RC_{Ry}-P_{zx}G_z]+[2(V_{xx}^R-V_{yy}^R)C_{Ry}-(V_{xy}^R+U_{xy}^R)F_{Rx} -Q_{zy}G_z][2(U_{zz}^R-V_{yy}^R)C_{Ry}-V_{xy}^RF_{Rx}-Q_{zy}G_z];$$

$$E_{56}^{(2)}(7)=\{[2(Y_{zz}^R-U_{xx}^R)F_{Rx}-V_{xy}^RC_{Ry}-P_{zx}G_z] \times[2(Y_{yy}^R-U_{xx}^R)F_{Rx}-(V_{xy}^R+Y_{xy}^R)C_{Ry}-P_{zx}G_z]+[2(Y_{zz}^R-V_{yy}^R)C_{Ry} -V_{xy}^RF_{Rx}-Q_{zy}G_z][2(W_{xx}^R-V_{yy}^R)C_{Ry}-(V_{xy}^R+Y_{xy}^R)F_{Rx}-Q_{zy}G_z]\}^{1/2}; \tag{31}$$

$$E_{78}^{(2)}(8)=\{[2(W_{zz}^R-U_{xx}^R)F_{Rx}-V_{xy}^RC_{Ry}-P_{zx}G_z] \times[2(W_{yy}^R-U_{xx}^R)F_{Rx}-(V_{xy}^R+W_{xy}^R)C_{Ry}-P_{zx}G_z]+[2(W_{zz}^R-V_{yy}^R)C_{Ry} -V_{xy}^RF_{Rx}-Q_{zy}G_z][2(Y_{xx}^R-V_{yy}^R)C_{Ry}-(V_{xy}^R+W_{xy}^R)F_{Rx}-Q_{zy}G_z]\}^{1/2}. \tag{32}$$

In the Γ_2 phase the two acoustic modes (Γ_{12} and Γ_{34}) are thus dynamically coupled with the RE subsystem [see Eqs. (28) and (30)].

We consider now the calculation of the tensor $\chi_{\alpha\beta}$ of the high-frequency magnetic susceptibility of orthoferrites. It follows directly from symmetry that the operators $\hat{\mu}_\xi^\alpha$ of the magnetic moments of the RE ions are connected with the operators of the effective spins $\hat{s}_\xi^\alpha$ by the relations

$$\hat{\mu}_\xi^\alpha=\mu_B\sum_\beta g_\xi^{\alpha\beta}\hat{s}_\xi^\beta, \tag{33}$$

$$g_\xi=\begin{pmatrix} g_{xx} & \pm g_{xy} & 0 \\ \pm g_{xy} & g_{yy} & 0 \\ 0 & 0 & g_{zz} \end{pmatrix}. \tag{34}$$

The plus sign corresponds to $\xi=5$ and 6 and the minus sign to $\xi=7$ and 8. At the same time, the g factor of the iron ions is isotropic and equal to 2. Thus, in the presence of a high-frequency magnetic field $\mathbf{h}(t)$ it is necessary to add to the Hamiltonian (3) a term

$$\hat{\mathcal{H}}(t)=-\mu_B h_x(t)[2\hat{F}_x+g_{xx}\hat{F}_{Rx}+g_{xy}\hat{C}_{Ry}] -\mu_B h_y(t)[2\hat{F}_y+g_{yy}\hat{F}_{Ry}+g_{xy}\hat{C}_{Rx}]-\mu_B h_z(t)[2\hat{F}_z+g_{zz}\hat{F}_{Rz}]. \tag{35}$$

After linearizing the equations of motion we now obtain a system of linear inhomogeneous equations, whose solution yields the components of the tensor $\chi_{\alpha\beta}$. The most important properties of the tensor $\chi_{\alpha\beta}$ are reflected in Table II, but its explicit form is too unwieldy to be written out here.

We point out in conclusion that the procedure of calculating and classifying the frequencies of homogeneous oscillations of a spin system, which was proposed in the present paper, can be used in the analysis of any multisublattice magnetic structure (antiferromagnetic or ferrimagnetic). It is also possible to consider the nuclear magnetic subsystem (in investigations of the dynamic frequency shift in NMR).

The authors thank I. E. Dzyaloshinskiĭ for a discussion of the work and for a number of useful remarks.

[1)] It is shown in Ref. 5 that for magnets having a noncollinear magnetic structure of exchange origin the number of acoustic modes is three, if the number of the sublattices exceeds two. In orthoferrites the number of acoustic modes is two, since their magnetic structure is collinear in the exchange approximation.

[2)] The resonant frequencies can be calculated also on the basis of the Landau–Lifshitz equations in which, however, it is necessary to change over from the sublattice magnetic-moment vectors to their irreducible linear combinations.

[1]I. E. Dzyaloshinskiĭ and B. T. Kukharenko, Zh. Eksp. Teor. Fiz. **70**, 2360 (1976) [Sov. Phys. JETP **43**, 1232 (1976)].
[2]A. F. Andreev and V. I. Marchenko, Zh. Eksp. Teor. Fiz. **70**, 1522 (1976) [Sov. Phys. JETP **43**, 794 (1976)].
[3]V. E. Naish and E. A. Turov, Fiz. Met. Metalloved. **11**, 161 (1961).
[4]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics) Fizmatgiz, 1963 [Pergamon, 1968].
[5]B. J. Halperin and W. M. Saslow, Phys. Rev. B **16**, 2154 (1977).
[6]A. Abragam and B. Bleaney, Electron paramagnetic resonance of transition ions. Clarendon Press, Oxford (1970).

Translated by J. G. Adashko

Oscillations of the thermoelectric power of cadmium in magnetic breakdown

V. I. Gostishchev, S. E. Dem'yanov, M. Glin'skiĭ, and A. A. Drozd

Institute of Solid State and Semiconductor Physics, Belorussian Academy of Sciences; International Laboratory for Strong Magnetic Fields and Low Temperatures, Wroclaw, Poland
(Submitted 25 October 1978)
Zh. Eksp. Teor. Fiz. **76**, 1392–1398 (April 1979)

The transverse thermoelectric power and the magnetoresistance were measured in cadmium of high purity, of orientation [10$\bar{1}$0] in fields up to 150 kOe. Oscillations of the thermoelectric power were observed in a wide range of angles when **H** was rotated in the (10$\bar{1}$0) plane. Coherent magnetic breakdown between the first and second bands of the Fermi surface is used to explain the results.

PACS numbers: 72.15.Jf, 72.15.Gd

Until recently, magnetic breakdown (MB) in cadmium was investigated mainly with the aid of the de Haas–van Alphen (dHvA) effect.[1,2] The studies of the Fermi surface of cadmium by Tsui and Stark[1] has shown that MB can occur between sheets of the Fermi surface of the first and second band through a gap of spin-orbit origin. Thus, the section of the monster in the ΓKM plane can take the form of single-, two-, and three-lobe figure in the case of MB near the point K. A unique "bridge" is provided in this case for the carrier by the sharp peak of the α-pocket in the first zone, which passes close to the K point but without touching it (Fig. 1). When H was rotated in the (11$\bar{2}$0) plane around the direction of the opening of the monster, new extremal sections γ' were observed in Ref. 1. They result from MB in the AHL plane between the pocket of the first band and the corrugated cylinder of the second, and branch away on the angular diagram from the usual sections of the monster γ. The combined two-band region which is produced because the cylinder is bounded from above and from below by pockets, has the observed extremal sections γ'.

However, a study of the MB phenomena by the dHvA method is a far from simple problem because of the complex spectrum of the observed frequencies and of the difficulty of its interpretation. From this point of view, the thermoelectric-power method, which has recently yielded a number of interesting results,[3-5] is preferable. It is shown that the thermoelectric-power coefficient S is sensitive to a restructuring of the electron energy spectrum by the magnetic breakdown, and depends little on the various scattering process in strong magnetic fields H. Compared with the magnetoresistance, whose oscillations are not observed in MB that leads to a change from open to closed trajectories,[6] a measurement of $S(H)$ makes it possible to observe such configurations in experiment. In addition, both the thermoelectric power and the magnetoresistance are oscillating functions of the magnetic field as a result of the Shubnikov–de Haas effect. To our knowledge, no such Shubnikov oscillations of the thermoelectric powers have observed so far, and the observed oscillations have been convincingly attributed to MB.

Cadmium remains perhaps the only hexagonal metal for which no investigations of oscillatory quantum phenomena were made by the thermoelectric-power method.

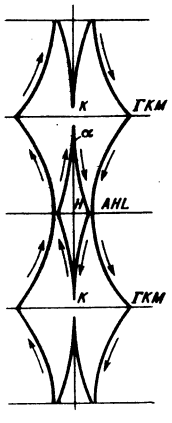

FIG. 1. Section of monster and pockets by the (11$\bar{2}$0) plane along the KH line. The arrows show the direction of electron motion.

 0038-5646/79/040708-05$2.40