Light scattering by magnons and magneto-optical effects in multisublattice magnets

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Light scattering by all possible branches of magnons with $k = 0$ in a multisublattice magnet is investigated. A procedure is proposed for analyzing the form of the scattering tensor, with account taken of the magnetic symmetry of the crystal. It is shown that the scattering tensor is connected with the magneto-optical constants of the crystal only when the scattering is by definite magnon branches. A procedure is developed for quantizing the Hamiltonian expressed in terms of irreducible combinations of spin operators; this procedure simplifies the calculation of the u–v coefficients in the case of a multisublattice magnet. It is established that the exchange branches of the magnon spectrum in antiferromagnets correspond predominantly to oscillations of the weak-antiferromagnetism vector. Light scattering by magnons in $ErFeO_3$ in various magnetically ordered phases is considered. The intensities are estimated and it is shown that as a rule the intensity of scattering by exchange magnon branches is less than by acoustic branches.

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INTRODUCTION

The spectrum of the spin waves (SW) in a multisublattice magnet contains besides acoustic branches also exchange branches that are the analog of optical phonons. These branches depend little weakly on the magnetic field and their frequencies can be estimated from the magnetic-ordering temperature.

Raman scattering of light (RSL) supplemented with experiments on antiferromagnetic resonance permits a study of all the spin-wave branches of the spectrum and its use to judge the magnetic structure of a crystal.

In the investigation of light scattering in magnets, interest attaches to knowledge of the form of the single-magnon scattering tensor. This analysis can be based on an allowance for the magnetic symmetry of the crystal, which determines its magnetic space group.[1] In the case of single-particle scattering of light by elementary excitations that have activation energy, when the momentum $k = \omega/c$ transferred to the spin waves is small ($k \sim 10^5$ cm^{-1} at $\omega \sim 10^{15}$ sec^{-1} $ak \sim 10^{-3}$, and a is the lattice constant), the spatial dispersion of these spin waves and of the Raman-scattering tensor can be neglected. It suffices therefore to use the magnetic point group for the analysis of the form of the RSL tensor.

It is known that the phase transition from the paramagnetic state to the magnetically ordered one is accompanied, as a rule, by magnetic structures whose symmetry is subgroup-coupled to the symmetry of the paramagnetic phase (PP).[2] The Hamiltonian of the magnetic subsystem of the crystal near the phase-transition point can then be constructed on the basis of the PP symmetry group.[3] If the exchange integrals depend little on the temperature and the striction distortions can be neglected, this Hamiltonian can be used to study the static and dynamic properties of the magnet also far from the phase transition. It is this traditional[3,4] procedure that we shall use in the present paper to analyze the form of the RSL tensor.

We denote by $\Delta\varepsilon_{ij}$ the spin-dependent part of the dielectric polarizability of the crystal. Usually $\Delta\varepsilon_{ij}$ is represented as a series in powers of the ion spins.[5] It is shown in Ref. 5 that for crystals with small ratio of the exchange interaction to the separation of the ground and lowest excited states of the ion it suffices to retain the first two terms of this expansion. We express $\Delta\varepsilon_{ij}$ in powers of irreducible operators $\mathscr{L}$, each a superposition of atomic-spin operators that transform in accord with irreducible representations (IR) of the PP group:

$$\Delta\varepsilon_{ij} = \sum a_{ijl}\mathscr{L}_l + \sum a_{ijlm}\mathscr{L}_l\mathscr{L}'_m \tag{1}$$

(the summation is over l, $\vec{\mathscr{L}}$, m, and $\vec{\mathscr{L}}'$). The indices l and m number the coordinate axes, while $\vec{\mathscr{L}}$ and $\vec{\mathscr{L}}'$ are the types of the irreducible operators. $\vec{\mathscr{L}}$ can be represented in the usual manner in the form of a sum $\vec{\mathscr{L}} = \vec{\mathscr{L}}_0 + \mathbf{L}$, in which $\vec{\mathscr{L}}_0$ are the equilibrium values determined by the magnetic ordering and $\mathbf{L}$ are small deviations due to the spin oscillations. If the $\vec{\mathscr{L}}$ are replaced in (1) by their equilibrium values $\vec{\mathscr{L}}_0$, we obtain the tensor

$$\Delta\varepsilon_{ij}^{(0)} = \sum a_{ijl}\mathscr{L}_{0l} + \sum a_{ijlm}\mathscr{L}_{0l}\mathscr{L}_{0m} \tag{2}$$

(summation over l, $\vec{\mathscr{L}}_0$, and m), where in the absence of absorption the first term describes the Faraday effect (FE) and the second the Cotton-Mouton effect (CME).

Allowance for the terms linear in the deviations from the equilibrium values of the operators $\mathbf{L}$ yields that part of the tensor $\Delta\varepsilon_{ij}^{(1)}$ which determines the single-magnon RSL

$$\Delta\varepsilon_{ij}^{(1)} = \sum a_{ijl}L_l + \sum a_{ijlm}L_l\mathscr{L}_{0m} \tag{3}$$

(summation over l, m, $\vec{\mathscr{L}}_0$, and $\mathbf{L}$).

The components of the single-magnon RSL are the matrix elements

$$\frac{1}{(n_\nu+1)^{1/2}}\langle n_\nu|\Delta\varepsilon_{ij}^{(1)}|n_\nu+1\rangle,$$

where n_ν are the magnon occupation numbers and the subscript ν labels the branches of the SW spectrum. The determination of the form of the RSL tensor reduces thus to an investigation of the forms of the tensors a_{ijl} and a_{ijlm} in (3). It must be recognized here that a_{ijl} has the same structure as the FE constants in (2), i.e., the presence of the time-reversal operation in the PP group should lead in the absence of absorption to the relations $a_{ijl} = -a_{jil}$.

The first and second terms of (3) are thus respectively antisymmetric and symmetric in the indices ij, and the tensor of the RSL by magnons is not purely antisymmetric (see also Ref. 6 on this subject). The components of the tensors a_{ijl} and a_{ijlm} in (3) differ from zero if the direct products

$$\Gamma(\mathrm{E})\times\Gamma^*(\mathrm{E})\times\Gamma(L_l^\nu), \tag{4}$$

$$\Gamma(\mathrm{E})\times\Gamma^*(\mathrm{E})\times\Gamma(L_l^\nu)\times\Gamma^*(\mathscr{L}_{0m}) \tag{5}$$

contain a unitary representation. Here $\Gamma(\mathrm{E})$ is the PP-group IR with respect to which the electric field-strength vector **E** is transformed, and $\Gamma(\mathscr{L}_{0m})$ are the IR with respect to which the equilibrium magnetic configurations are transformed; $\Gamma(L_l^\nu)$ are the IR of the irreducible operators of the spin flops of the branch ν.

To analyze further the form of the RSL tensor we must recognize that in the oscillations of the magnon mode ν there can participate the irreducible spin flop operators **L** which transform in accord with a single corepresentation of the magnetic group of the crystal, but generally speaking in accord with different IR of the PP group.[4,7] It can be shown here that the direct product of the PP-group IR in accord with which the irreducible spin-flop operators corresponding to one magnon mode are transformed, always contain IR of an equilibrium magnetic configuration. This condition, with account taken of relations (4) and (5), solves the problem of finding the form of the tensor of RSL by magnons in various magnetically ordered phases.

From (4) and (5) one can determine the structures of the FE and CME tensors in (2) when $\Gamma(L_l^\nu)$ is replaced by IR of the equilibrium configurations $\Gamma(\mathscr{L}_{0m})$.[8] Thus, for example, it follows from (4) that if a transition into a magnetically ordered phase occurred in accord with an IR with $\mathbf{k}\neq 0$ (this case includes, e.g., doubling of the crystal-chemical cell or the onset of certain inhomogeneous structures), the direct product cited will not contain a unitary representation, so that there are no linear magneto-optical effects without a magnetic field.

It follows from (4) and (5) that the components of the RSL tensors (3) are connected with the FE and CME constants that enter in (2) for only those modes in which a part is played by the irreducible operators that transform in accord with IR that coincide with the IR of the equilibrium configurations $\mathscr{L}_0$.

The magneto-optical constants that determine the FE and CME in multisublattice magnets contain a large number of contributions from different magnetic configurations. Using the cited condition for the connection between the RSL tensor components and the FE and CME constants, as well as the fact that these contributions enter in the RSL tensor with different multipliers determined by the coefficients of the u–v transformation of the spin-system Hamiltonian, we can distinguish between these contributions by comparing the results of the measurements of the absolute values of the scattering intensities with the magneto-optic measurements.

We demonstrate in this paper, with orthoferrites as the example, that a consistent application of symmetry considerations allows us not only to propose a simple procedure for calculating the spectrum of the homogeneous magnetization oscillations,[4] but also to calculate the u–v transformation matrix that describes the transition to normal magnon modes. Naturally, this rigorous approach, which makes no use of a subgroup coupling of the ground state of a magnetic crystal with the PP, must be based on the use of the magnetic-symmetry group at the crystal when calculating both the frequencies of the SW (Ref. 7) and their polarizations (of the u–v transformation matrix). After this paper was already written, such an approach was proposed in Ref. 9, where the SW spectrum was calculated with wave vectors along symmetry directions of the Brillouin zone for Mn_3NiN.

On the basis of our proposed relations (4) and (5) we obtained the structure of the RLS tensor in four-sublattice AF with symmetry group D_{2h}^{16}, covering a large class of magnets such as orthoferrites.

We investigated the behavior of the light-scattering intensity for all branches of the SW spectrum in the region of spin-flip phase transitions.

1. CALCULATION OF THE AMPLITUDES OF NORMAL MAGNON MODES IN AN ORTHOFERRITE

From the symmetry considerations used in Ref. 4 to analyze the frequencies of homogeneous magnon modes it follows that the dispersion equation breaks up into individual blocks corresponding to oscillations of like symmetry. Besides the natural-oscillation frequencies, a fundamental role is played in the study of the high-frequency properties of magnets, and particularly of RSL, by the amplitudes of the normal oscillations of the magnetization, which are determined by the u–v transformation coefficients. Let us show that symmetry considerations simplify also the procedure of calculating the u–v coefficient matrix that describes a transition from irreducible spin-flop operators to normal magnon modes. We shall illustrate these general considerations using as a particular example the calculations of the u–v coefficients at $k=0$ in the four-sublattice antiferromagnet $ErFeO_3$.

The crystal magnetic-subsystem Hamiltonian that describes the ground states and the uniform oscillations of the magnetizations can be written, following Ref. 4, in the form

$$\begin{aligned}H=&J_{4x}A_x^2+J_{3y}G_y^2+J_{2z}C_z^2+D_{1z}A_xG_y+D_{1y}A_xC_z\\&+D_{1x}G_yC_z+J_{1x}F_x^2+J_{2y}C_y^2+J_{3z}G_z^2+D_{2z}F_xC_y+D_{2y}F_xG_z\\&+D_{2x}C_yG_z+J_{2x}C_x^2+J_{1y}F_y^2+J_{4z}A_z^2+D_{3z}C_xF_y\\&+D_{3y}C_xA_z+D_{3x}F_yA_z+J_{3x}G_x^2+J_{4y}A_y^2+J_{1z}F_z^2\\&+D_{4z}G_xA_y+D_{4y}G_xF_z+D_{4x}A_yF_z+e_1G_x^4+e_2G_x^2G_z^2+e_3G_z^4.\end{aligned} \tag{6}$$

To describe the spin-flip transition in $ErFeO_3$, we included in (6) terms proportional to the four power of the magnetization; J, D, and e are respectively the exchange, Dzyaloshinskiĭ, and anisotropy constants. Also connected with the anisotropy energy are the differences between J_{4x} and J_{4y}, J_{3x} and J_{3y}, etc. The irreducible operators **F**, **G**, **C**, and **A** in (6), as well as their classification in accord with the IR of the PP group D_{2h}^{16}, are given according to Ref. 10 by

$$\begin{array}{l|l|l} \Gamma_1 & A_xG_yC_z & \mathbf{F}=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3+\mathbf{S}_4, \\ \Gamma_2 & F_xC_yG_z & \mathbf{G}=\mathbf{S}_1-\mathbf{S}_2+\mathbf{S}_3-\mathbf{S}_4, \\ \Gamma_3 & C_xF_yA_z & \mathbf{C}=\mathbf{S}_1+\mathbf{S}_2-\mathbf{S}_3-\mathbf{S}_4, \\ \Gamma_4 & G_xA_yF_z & \mathbf{A}=\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3+\mathbf{S}_4. \end{array} \tag{7}$$

We shall consider only the temperature region in which there is no ordering of the magnetic subsystem of the rare-earth ions. We take its influence into account in the usual manner,[11] assuming that the constants of the Hamiltonian (6) are renormalized by the Fe–R interaction and are thus temperature-dependent. Since a G-type ordering is realized in orthoferrites, i.e., **G** is the largest antiferromagnetism vector, only terms containing **G** are retained in the fourth-order anisotropy.

It is known that at temperatures T_1 and T_2 two second-order phase transitions take place in $ErFeO_3$ (see Ref. 11 for details). At $T\leqslant T_1$ a magnetically ordered phase is realized in which the equilibrium values F_{0x}, G_{0z} and C_{0y} corresponding to the IR Γ_2 differ from zero. We shall call this phase Γ_2 and classify in similar manner the phases considered later. In the interval $T_1<T<T_2$ is realized a phase Γ_{24} in which the vector **G** rotates in the xz plane and the nonvanishing quantities are the equilibrium values of the irreducible operators that transform in accord with the Γ_2 and Γ_4 IR. The phase realized above T_2 is Γ_4 with equilibrium values G_{0x}, A_{0y}, F_{0z}.

We consider now the procedure of second quantization of the Hamiltonian (6). We express the operators **L** in terms of the operators $\mathbf{S}^{\prime(\alpha)}$ expressed in a local coordinate frame with z' axis directed along the equilibrium value of the spin $\mathbf{S}_0^{(\alpha)}$. The operators $\mathbf{S}^{(\alpha)}$ in the crystal coordinate frame are connected with the operators in the local frame by the relation

$$S_i^{(\alpha)}=p_{ij}^{(\alpha)}S_j^{\prime(\alpha)}, \tag{8}$$

and the index α numbers the positions of the Fe^{3+} ions.

The matrices $\hat{p}^{(\alpha)}$ for the ion positions that are interchanged by symmetry operations are not independent. Thus, if the equilibrium value fo $\mathbf{S}_0^{(\beta)}$ is obtained from $\mathbf{S}_0^{(\alpha)}$ with the aid of the magnetic-group symmetry operation g, we have

$$\hat{p}^{(\beta)}=\hat{g}\hat{p}^{(\alpha)}, \tag{9}$$

where $\hat{g}$ is the g transformation matrix (we assume that the unit vectors of the local frame transform as axial t-odd vectors). Because of this we can find the connection between the operators **L** expressed in the crystal coordinate frame and $\vec{\mathbf{L}}'$ defined by relations (7), in which the operators $\mathbf{S}^{(\alpha)}$ must be replaced by $S^{\prime(\alpha)}$. For example, in the phase Γ_4 we have

$$\begin{pmatrix}A_x\\G_y\\C_z\end{pmatrix}=\hat{p}^{(1)}\begin{pmatrix}F_x'\\C_y'\\C_z'\end{pmatrix},\quad \begin{pmatrix}G_x\\A_y\\F_z\end{pmatrix}=\hat{p}^{(1)}\begin{pmatrix}C_x'\\F_y'\\F_z'\end{pmatrix},$$
$$\begin{pmatrix}F_x\\C_y\\G_z\end{pmatrix}=\hat{p}^{(1)}\begin{pmatrix}A_x'\\G_y'\\G_z'\end{pmatrix},\quad \begin{pmatrix}C_x\\F_y\\A_z\end{pmatrix}=\hat{p}^{(1)}\begin{pmatrix}G_x'\\A_y'\\A_z'\end{pmatrix}, \tag{10}$$

where the matrix $\hat{p}^{(1)}$ defined for the ion Fe^{3+} in the position 1 with the aid of (8) is of the form

$$\hat{p}^{(1)}=\begin{pmatrix}-\sin\varphi & \cos\chi\cos\varphi & \sin\chi\cos\varphi\\ \cos\varphi & -\cos\chi\sin\varphi & \sin\chi\sin\varphi\\ 0 & \sin\chi & \cos\chi\end{pmatrix}; \tag{11}$$

here

$$\sin\chi\cos\varphi=G_{0x}(4S)^{-1},\quad \sin\chi\sin\varphi=A_{0y}(4S)^{-1},$$
$$\cos\chi=F_{0z}(4S)^{-1},$$

where S is the ion spin. We note that in the phase Γ_4 the angle φ is small and χ is close to $\pi/2$. In the derivation of (10) it must be taken into account that some of the operations g transform left-hand systems into right-hand.

The operators $\mathbf{L}'$ are determined by the spin operators in the local coordinate frame and are therefore directly connected with the Holstein-Primakoff spin-flops a_{α^+} and a_α. We introduce linear combinations of the operators a_α ($\alpha=1,2,3,4$), having the same structure as the irreducible operators (7):

$$a_F=\tfrac{1}{2}(a_1+a_2+a_3+a_4),\quad a_G=\tfrac{1}{2}(a_1-a_2+a_3-a_4),$$
$$a_C=\tfrac{1}{2}(a_1+a_2-a_3-a_4),\quad a_A=\tfrac{1}{2}(a_1-a_2-a_3+a_4). \tag{12}$$

It can be easily seen that the operators a_L, a_L^+ ($L=F,G,C,A$) satisfy the usual commutation relations

$$[a_L,a_{L'}]=[a_L{}^+,a_{L'}{}^+]=0,\quad [a_L,a_{L'}{}^+]=\delta_{L,L'}. \tag{13}$$

Thus, using the Holstein-Primakoff representation, we obtain with the aid of (7) and (12)

$$L'^+=(8S)^{1/2}a_L,\quad L'^-=(8S)^{1/2}a_L{}^+,\quad L'^\pm=L_x'\pm iL_y', \tag{14}$$

$$F_z'=4S-(a_F{}^+a_F+a_G{}^+a_G+a_C{}^+a_C+a_A{}^+a_A),$$
$$C_z'=-(a_F{}^+a_C+a_G{}^+a_A+\text{э.с.}),\quad G_z'=-(a_F{}^+a_G+a_C{}^+a_A+\text{H.c.}),$$
$$A_z'=-(a_F{}^+a_A+a_C{}^+a_G+\text{H.c.}). \tag{15}$$

Using (10)–(15) we can rewrite the Hamiltonian (6) in terms of the operators a_L and a_L^+.

The ground state of the system, as usual, is determined from the condition that the coefficients of the terms linear in the Bose operators a_L and a_{L^+} vanish. It is easily found that following the transformation of (6) with the aid of (10), followed by (14) and (15), that part of the Hamiltonian which is quadratic in these Bose operators contains no products of the form a_Fa_G, a_Fa_A, a_Ca_G, and a_Ca_A. The absence of these terms is not accidental, but is due to carrying out the second quantization (6) in terms of irreducible operators.

As a result, the matrix for the u–v coefficients introduced in the usual manner

$$a_L=\sum_\nu\{u_{L\nu}\xi_\nu+v_{L\nu}{}^*\xi_\nu{}^+\}, \tag{16}$$

breaks up, say in the Γ_4 phase, into two independent blocks connected respectively with the operators a_Fa_C and a_Ga_A. Here ξ_ν^+ and ξ_ν are the creation and annihilation operators of the magnons of the branch ν at $\mathbf{k}=0$.

Thus, the use of irreducible operators makes it possible to retain the advantages of the symmetry approach not only in the calculation of the magnon frequencies,[4] but also in the

TABLE I.

	$\overset{+}{\nu}_{14}$	$\overset{-}{\nu}_{14}$		$\overset{+}{\nu}_{23}$	$\overset{-}{\nu}_{23}$
$d_{F\mu}$	$-i\dfrac{D_{1x}}{E_{2zx}}\left(\dfrac{E_{2zx}}{E_{4yx}}\right)^{1/4}$	$i\left(\dfrac{\overset{-}{\nu}_{14}}{E_{1zx}}\right)^{1/2}$	$d_{G\mu}$	$i\dfrac{D_{2x}E_{1yx}}{E_{4zx}^{3/4}E_{2yx}^{5/4}}$	$-i\left(\dfrac{E_{1yx}}{\overset{-}{\nu}_{23}}\right)^{1/2}$
$t_{F\mu}$	$-i\dfrac{D_{1x}E_{1zx}}{E_{4yx}^{3/4}E_{2zx}^{5/4}}$	$i\left(\dfrac{E_{1zx}}{\overset{-}{\nu}_{14}}\right)^{1/2}$	$t_{G\mu}$	$i\dfrac{D_{2x}}{E_{2yx}}\left(\dfrac{E_{2yx}}{E_{4zx}}\right)^{1/4}$	$-i\left(\dfrac{\overset{-}{\nu}_{23}}{E_{1yx}}\right)^{1/2}$
$t_{C\mu}$	$\left(\dfrac{E_{2zx}}{E_{4yx}}\right)^{1/4}$	$\dfrac{D_{1x}(E_{1zx}\overset{-}{\nu}_{14})^{1/2}}{E_{2zx}E_{4yx}}$	$t_{A\mu}$	$\left(\dfrac{E_{4zx}}{E_{2yx}}\right)^{1/4}$	$\dfrac{D_{2x}}{E_{2yx}}\left(\dfrac{E_{1yx}}{\overset{-}{\nu}_{23}}\right)^{1/2}$
$d_{C\mu}$	$\left(\dfrac{E_{4yx}}{E_{2zx}}\right)^{1/4}$	$\dfrac{D_{1x}}{E_{2zx}}\left(\dfrac{E_{1zx}}{\overset{-}{\nu}_{14}}\right)^{1/2}$	$d_{A\mu}$	$\left(\dfrac{E_{2yx}}{E_{4zx}}\right)^{1/4}$	$\dfrac{D_{2x}}{E_{2yx}}\dfrac{(E_{1yx}\overset{-}{\nu}_{23})^{1/2}}{E_{4zx}}$

calculation of the u–v coefficients for arbitrary multisublattice magnets.

We introduce the quantities

$$t_{L\nu}=u_{L\nu}+v_{L\nu},\quad d_{L\nu}=u_{L\nu}-v_{L\nu}, \tag{17}$$

which we shall need later on and which satisfy the usual normalization conditions

$$\sum_{\nu}(t_{L\nu}d^{*}_{L'\nu}+t^{*}_{L'\nu}d_{L\nu})=2\delta_{L,L'},$$

$$\sum_{L}(t_{L\nu}d^{*}_{L\nu'}+t^{*}_{L\nu'}d_{L\nu})=2\delta_{\nu\nu'}.$$

The expressions for t and d in the Γ_4 phase are gathered in the table. [We have introduced here the notation $E_{\alpha ij}=2(J_{\alpha i}-J_{3j})$. The " + " and " − " signs refer respectively to the exchange and acoustic modes. The expressions for the frequencies of these modes in the phases Γ_1, Γ_2, and Γ_4 are given in Ref. 4. The connection between our notation for the frequencies, e.g., ν_{14}, and that of Ref. 4 (E_{14}), is described by the relation $\nu_{14}=E_{14}(4M_0)^{-1}$]. In the calculation of the u–v coefficients, just as in the calculations of the frequencies in Ref. 4, we used the approximations

$$|J_{\alpha i}|\gg D\gg|J_{\alpha i}-J_{\alpha j}|,\quad \alpha=1,\ 2,\ 3,\ 4;\quad ij=x,\ y,\ z. \tag{18}$$

The values of E in the table are determined by the exchange constants, $E\sim\mu H_E$. The frequencies of the acoustic modes of the spectrum are known to be of the order of $\nu^{-}\sim\mu(H_EH_A)^{1/2}$, while the frequencies of the exchange modes are $\nu^{+}\sim\mu H_E$, where H_E is the exchange field and H_A the anisotropy field. Therefore the maximum value of the coefficient t or d for the acoustic branch is of the order of $(H_E/H_A)^{1/4}$, i.e., exchange enhancement takes place, whereas for the exchange branch the value is $(H_E/H_{E'})^{1/4}\sim 1$ and there is no exchange enhancement. Substituting the solutions for the u–v coefficients in (14) and then changing over to (10) we can see that the oscillations corresponding to small deviations of the antiferromagnetism vector G are always exchange-enhanced for the acoustic branches.

In the case of the exchange branches, the u–v coefficients with maximum value of the order of unity enter in the formulas that describe the oscillations of the weak antiferromagnetism vectors **A** and **C**, whereas the equations for the oscillations of the F and G types contain u–v coefficients smaller by a factor D/J. The hierarchy of the u–v coefficients for the exchange modes reflects the possibility of a transition from the four-sublattice model of the orthoferrite to a two-sublattice model. In this transition, the exchange branches do not contribute to the oscillations of the vectors F and G. Thus, the exchange branches of the spectrum in multisublattice AFM correspond predominantly to oscillations of the weak-antiferromagnetism vectors.

We consider now the spin-flip region—the Γ_{24} phase. In this phase, the following equilibrium configurations differ simultaneously from zero:

$$\begin{gathered}\bar{F}_x\approx F_{0x}\sin\theta,\quad \bar{C}_y\approx C_{0y}\sin\theta,\\ \bar{G}_z\approx 4S\sin\theta,\\ \bar{G}_x\approx 4S\cos\theta,\quad \bar{A}_y\approx A_{0y}\cos\theta,\\ \bar{F}_z\approx F_{0z}\cos\theta,\end{gathered} \tag{19}$$

where F_{0x}, C_{0y} and F_{0z}, A_{0y} are the equilibrium values in the respective phases Γ_2 and Γ_4 and are defined in Ref. 4. The angle between the vector **G** and the x axis of the crystal is given by the relation[11]

$$\sin\theta=\left(\frac{T_2-T}{T_2-T_1}\right)^{1/2}. \tag{20}$$

The expression for the SW frequencies in the Γ_{24} phase are given in the Appendix.

The behavior of the branches of the spectrum in the spin-flip region are shown schematically in the figure. At $T=T_1$ and $T=T_2$ the equations for $\nu_{\rm II}$, $\nu_{\rm III}$, and $\nu_{\rm IV}$ go over into those for the frequencies in Ref. 4, with allowance for the renormalization of all the quantities of this reference on account of the interaction with the rare-earth sublattice. From the formulas given in the Appendix for the frequencies it can be seen that ν_I vanishes at $T=T_1$ and $T=T_2$. The reason is that the frequencies were calculated without allowance, e.g., for the magnetoelastic interaction that leads to the appearance of a gap in the spectrum of the softening magnon mode. Such a calculation for the two-sublattice model of the orthoferrite was carried out in Ref. 12. We have calculated the u–v coefficients in the Γ_{24} phase, but in view of their length we present the results only for the soft mode:

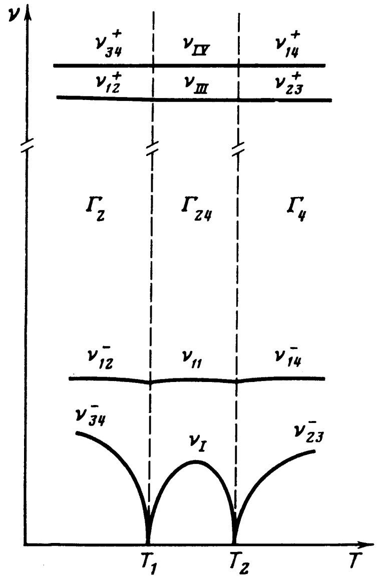

FIG. 1.

$$t_F=0,\quad d_F=\frac{D_{2x}D_{4z}}{E_2E_4}\left(\frac{E_1}{K_4}\right)^{1/4}(\sin\theta\cos\theta)^{1/2},\quad d_A=t_A\frac{\nu_{\rm I}}{E_4},$$

$$t_A=i\frac{D_{2x}}{E_2}\left(\frac{E_1}{K_4}\right)^{1/4}\left(\frac{\cos\theta}{\sin\theta}\right)^{1/2},\quad d_C=-\left(\frac{K_4}{E_1}\right)^{1/4}(\sin\theta\cos\theta)^{1/2},$$

$$t_C=\frac{D_{4z}}{E_4}\left(\frac{E_1}{K_4}\right)^{1/4}\left(\frac{\sin\theta}{\cos\theta}\right)^{1/2}, \tag{21}$$

$$t_G=-\left(\frac{K_4}{E_1}\right)^{1/4}(\sin\theta\cos\theta)^{1/2},\quad d_G=t_G^{-1};$$

the quantities E_α ($\alpha=1,2,3,4$) and K_4 are defined in the Appendix. The coefficients t_A, t_C, and d_G have singularities at the phase-transition points T_1 and T_2. These singularities, as will be shown below, lead to an abrupt increase of the intensity of the Raman scattering of light in the vicinity of the phase transitions.[1]

2. MAGNETO-OPTICAL EFFECTS AND BEHAVIOR OF THE SCATTERING INTENSITIES

Since the quantities G_{0x}, A_{0y} and F_{0z} differ from zero, the FE is described according to (2) and (4) by the component

$$\Delta\varepsilon_{xy}^{(0)}=\lambda_1 G_{0x}+\lambda_2 A_{0y}+\lambda_3 F_{0z}, \tag{22}$$

where the quantities λ_1, λ_2, and λ_3 describe the contribution made to the FE by the antiferromagnetism, weak antiferromagnetism, and magnetization vectors, respectively. The nonvanishing quantities in the Γ_2 phase are F_{0x}, C_{0y}, G_{0z}. Therefore the FE is described by the dielectric-constant component

$$\Delta\varepsilon_{yz}^{(0)}=\lambda_1' G_{0z}+\lambda_2' C_{0y}+\lambda_3' F_{0z}. \tag{23}$$

There is no Faraday effect in the magnetically ordered phase Γ_1.

Here and elsewhere we shall not write out the invariants connected with the rare-earth sublattice.

Using (5), we can obtain expressions for the CME constants. In the Γ_4 phase these constants are of the form

$$\Delta\varepsilon_{ii}^{(0)}=\lambda_5^{(i)} G_{0x}^2+\lambda_6^{(i)} A_{0y}^2+\lambda_7^{(i)} F_{0z}^2+\lambda_8^{(i)} G_{0x}A_{0y}+\lambda_9^{(i)} G_{0x}F_{0z}+\lambda_{10}^{(i)} A_{0y}F_{0z}. \tag{24}$$

The form of the CME constants for the phases Γ_1 and Γ_2 are easily obtained from (5), but we shall not present them here.

We note the off-diagonal (anisotropic) dielectric-constant components connected with the CME are absent in all the phases Γ_1, Γ_2, and Γ_3.[2] In the Γ_{24} phase in the spin-flip region, however, an off-diagonal symmetric component appears,

$$\Delta\varepsilon_{xz}^{(0)}=\delta_1\bar{G}_x\bar{G}_z+\delta_2\bar{G}_x\bar{F}_x+\delta_3\bar{G}_x\bar{C}_y, \tag{25}$$

and its value varies with temperature like $\Delta\varepsilon_{xz}\sim\sin\theta\cos\theta$. Application of a magnetic field whose components are transformed in accordance with IR that do not coincide with the IR of the equilibrium magnetic configurations leads to the appearance of mixed phases and consequently of anisotropic symmetric components of the dielectric constant. In particular, as can be seen from (5), an invariant is possible, containing the antiferromagnetism vector and the field **H**. Because of this effect, antiferromagnetic domains were observed[13] in dysprosium orthoferrite in the Γ_1 phase following application of a field $\mathbf{H}\|z$ ($\Gamma(H_z)=\Gamma_4$). The CME constants of the orthoferrites are given in the same reference with account taken of equilibrium configuration of type F and G only.

We consider now the form of the tensors of RSL by magnons in the magnetically ordered phase Γ_4. In this phase a part is played in the oscillations of the exchange mode ν_{14}^+ and of the acoustic mode ν_{14}^- by irreducible spin-flop operators that transform in accord with IR of Γ_1 and Γ_4. Similarly, for the modes ν_{23}^- and ν_{23}^+ the pertinent IR are of Γ_2 and Γ_3.[3] Therefore, as follows from (4) and (5), the RSL tensor in the Γ_4 phase is of the form

$$a_{ij}(\nu_{14}^\pm)=\begin{vmatrix} a_1^\pm & a_4^\pm & 0\\ a_5^\pm & a_2^\pm & 0\\ 0 & 0 & a_3^\pm\end{vmatrix},\quad a_{ij}(\nu_{23}^\pm)=\begin{vmatrix} 0 & 0 & a_6^\pm\\ 0 & 0 & a_7^\pm\\ a_8^\pm & a_9^\pm & 0\end{vmatrix}. \tag{26}$$

In the Γ_2 phase at the frequencies $\nu_{12}^\pm$ and $\nu_{34}^\pm$ the situation realized is similar to that just considered, and the RSL tensors are of the form

$$a_{ij}(\nu_{12}^\pm)=\begin{vmatrix} b_1^\pm & 0 & 0\\ 0 & b_2^\pm & b_4^\pm\\ 0 & b_5^\pm & b_3^\pm\end{vmatrix},\quad a_{ij}(\nu_{34}^\pm)=\begin{vmatrix} 0 & b_6^\pm & b_7^\pm\\ b_8^\pm & 0 & 0\\ b_5^\pm & 0 & 0\end{vmatrix}. \tag{27}$$

As noted earlier, the components of the RSL tensors are connected with the FE and CME constants only for those frequencies whose spin-flop-operator irreducible representations coincide with the irreducible representations of the equilibrium configurations. Therefore the components a_{ij} (ν_{23}) in the Γ_4 phase and a_{ij} (ν_{34}) in the Γ_2 phase do not contain the constants of the magneto-optical effects of these phases.

On the other hand, the diagonal components of the tensors a_{ij} (ν_{14}) and a_{ij} (ν_{12}) are connected with the constants of the quadratic magneto-optical effects. Obviously, the tensor for scattering by the SW modes corresponding to the operators **L**, which transform in accordance with the IR of the equilibrium configurations, will always have diagonal components. Let us examine in greater detail the structure of the components of the RSL tensors. By way of example, we determine the component $a_4^{\pm}$ of the tensor a_{ij} ($\nu_{14}^{\pm}$), which contains magneto-optical constants in the Γ_4 phase. In expression (3), this component corresponds to

$$\Delta\varepsilon_{xy}^{(1)}=\sum a_{xyl}L_l+\sum a_{xypm}L_p\mathscr{L}_{0m} \tag{28}$$

(summation over l, **L**, $\vec{\mathscr{L}}$, p, and m). According to (4) and (5), the antisymmetric part of (28), which is determined by the first term, contains the irreducible operators L_1 that transform in accord with $\Gamma_4(G_x, A_y, F_z)$. The symmetrical part of (28) (the second term) contains the operators L_p, which transform in accord with Γ_1 (A_x, G_y, C_z). The necessary next step is a transformation from the operators L_1 and L_p to the SW creation and annihilation operators in accord with Eqs. (10)–(16). Next, taking the matrix element of (28) [see the explanation of Eq. (3)], we obtain for $a_4^{\pm}$

$$\begin{aligned}a_4^{\pm}&=\lambda_1[iF_{0z}G_{0x}^{-1}d_{F\pm}-A_{0y}G_{0x}^{-1}t_{C\pm}]+\lambda_2t_{C\pm}-i\lambda_3d_{F\pm}\\&+(\sigma_1G_{0x}+\sigma_4A_{0y}+\sigma_7F_{0z})t_{F\pm}-(\sigma_3G_{0x}+\sigma_6A_{0y}+\sigma_9F_{0z})d_{C\pm}\\&+(\sigma_2G_{0x}+\sigma_5A_{0y}+\sigma_8F_{0z})[iF_{0z}G_{0x}^{-1}d_{C\pm}-A_{0y}G_{0x}^{-1}t_{F\pm}].\end{aligned} \tag{29}$$

We have left out here the terms $A_{0y}F_{0z}G_{0x}^{-2}\sim10^{-4}$ and introduced the notation $\sigma_1=a_{xyxx}$, $\sigma_2=a_{xyxy}$, $\sigma_3=a_{xyxz}$, etc.

The terms proportional to σ do not describe the magneto-optical effects in the Γ_4 phase. It can be easily seen that the symmetric terms connected with σ_i always enter in the off-diagonal components of the RSL tensors in orthoferrite phases Γ_4 and Γ_2. To compare the intensities of the scattering by the exchange and acoustic modes $\nu_{14}^{\pm}$, determined by the square of the modulus of the corresponding component $a_4^{\pm}$, definite assumptions must be made concerning the values of the constants λ_i and σ_i. Since we are taking into account the contribution made to $\Delta\varepsilon_{ij}^{(1)}$ by the spin-orbit scattering mechanisms, the constants σ are of second order of smallness in the spin-orbit interaction, while λ are of first order of smallness. Therefore $|\lambda|\gg|\sigma|$. Investigation[14] of the contribution of the antiferromagnetic and ferromagnetic vectors to the FE has established that λ_1 of the yttrium orthoferite is smaller than λ_3 by one or two orders of magnitude. We shall assume that $\lambda_1\sim\lambda_2\sim\lambda_3\cdot10^{-1}$ Using the values listed in the table for t and d, as well as the fact that the estimate $A_{0y}G_{0x}^{-1}\sim F_{0z}G_{0x}^{-1}\sim10^{-2}$ holds for the Γ_4 phase,[15] we obtain for the components a_4^+ and a_4^- from (29) the expressions

$$a_4^-\approx\lambda_3\left(\frac{\nu_{14}^-}{E_{1zx}}\right)^{1/2},\qquad a_4^+\approx\lambda_2\left(\frac{E_{2zx}}{E_{4yx}}\right)^{1/4}.$$

Let us compare the quantities a_4^+ and a_4^-, which determine the intensities of RSL by exchange and acoustic magnons. According to Ref. 16, for erbium orthoferrite $(\nu_{14}-E_{1zx}{}^{-1})^{1/2}\sim10^{-1}$. Taking $(E_{2zx}E_{4yx}{}^{-1})^{1/4}\sim1$, we obtain

$$a_4^+(a_4^-)^{-1}\sim10\lambda_2\lambda_3^{-1}\sim1.$$

If we assume now that $\lambda_3\gg\lambda_1,\lambda_2$, i.e., take into account only the ferromagnetic contribution to the FE, we get

$$a_4^+(a_4^-)^{-1}\sim10^{-1}.$$

The intensity of scattering by the exchange mode is thus appreciably less than for the acoustic mode. It must be noted, however, that this assumption may not hold in noncollinear magnetic structures, in which the equilibrium directions of the spin may not coincide with the orbital-momentum quantization axis determined by the crystal field.

For RSL by acoustic magnons in antiferromagnets one can identify scattering-tensor components that are exchange-enhanced. It is necessary for this purpose to find the IR according to which small deviations from the antiferromagnetism vector from the equilibrium position are transformed, inasmuch as for these IR the u–v components are a maximum. Knowing these IR, we can determine with the aid of (4) and (5) the corresponding components of the tensors a_{ijl} and a_{ijlm}. For example, in the Γ_4 phase small deviations of the antiferromagnetism vector G_y and G_z are transformed in accordance with Γ_1 and Γ_2, and consequently the symmetrical parts of the components a_4^-, a_5^- and a_6^-, a_8^- as well as the antisymmetric parts of the components $a_{7,9}^-$ will be exchange-enhanced.

The exchange-enhancement effects increase as the spin-flip points are approached, owing to the decrease of the frequency of the soft magnon mode. The symmetry of the new phase is determined by the orientation of the antiferromagnetism vector, and the vanishing of the frequency is due to the vanishing of the anisotropy that maintains the vector **G** in the previous position. Therefore the softened acoustic mode will be the one in whose oscillations participate irreducible operators that transform in accord with the *IR* of the new phase. For example, when the T_2 point is approached, the mode ν_{23}^- is softened in the Γ_4 phase. Similarly, in the magnetically ordered Γ_2 phase, the mode ν_{34}^- is softened when the point T_1 is approached and, according to (27), the antisymmetric parts of the components $b_{6,8}^-$ and the symmetric parts of the components $b_{7,9}^-$ increase in proportion to $(E_{1xz}/\nu_{34}^-)^{1/2}$, whereas the antisymmetric part of $b_{7,9}^-$ and the symmetric part of $b_{6,8}^-$ decrease in proportion to $(\nu_{34}-E_{1xz}{}^{-1})^{1/2}$. Since the Ginzburg-Levanyuk numbers of orthoferrites is small, $\mathrm{Gi}\sim10^{-8}$ (Ref. 11), the critical fluctuations become essential only in a very narrow region near the temperatures T_1 and T_2 of the transitions, and we do not consider them.

The tensor of scattering by a non-softening acoustic mode will contain the magneto-optical constants of the phase Γ_2. The off-diagonal part of the component $b_{4,5}^-$, which includes the constants λ_i' from (23), is exchange-weakened, just as the component $a_{4,5}^-$ which contains λ_i in the Γ_4 phase. However, the line corresponding to scattering by the mode ν_{12}^- in the Γ_2 phase was not observed in experiments on Raman scattering (RS) by magnons in $ErFeO_3$.[16] Since the coefficients preceding the mangneto-optical constants λ_i and λ_i' in the RS tensors are of the same order of magnitude, it can be concluded from the fact that no scattering by the mode ν_{12}^- is observed in the Γ_2 phase but does take place in the Γ_4 phase, that $\lambda_i'\ll\lambda_i$. The FE effect in the Γ_2 phase of $ErFeO_3$ should therefore be smaller than in the Γ_4 phase.

In the spin-flip region in the Γ_{24} phase the oscillations of all the modes are transformed in accord with the representations of Γ_{1234}. All the components in the scattering tensors differ therefore from zero. Using (20) and (21), let us examine the behavior of the intensity of scattering by the soft mode in the temperature region $T_1 < T < T_2$. The antisymmetric part of the xy and yx components in this temperature region behaves as $(T_2 - T)^{1/4}/(T - T_1)^{1/4}$. These components have thus singularities at $T = T_1$ and vanish at the point $T = T_2$ in accord with the experimental data of Ref. 16. The antisymmetric part of the yz and zy components is proportional to $(T - T_1)^{1/4}/(T_2 - T)^{1/4}$, i.e., has a singularity at $T = T_2$. In addition, a singularity in the spin-flip points will be possessed by the symmetric part of the yz and zy components:

$$a_{yz}^{(s)}(\nu_{\mathrm{I}}) \sim \mathrm{const}/(T_2-T)^{1/4} + \mathrm{const}/(T-T_1)^{1/4}.$$

The diagonal component of the scattering tensors will have no singularities whatever.

On the basis of (19) it is easy to obtain the temperature dependence, in the Γ_{24} phase, of the tensor components of the nonsoftening acoustic mode and of the exchange branches. This dependence should cause the scattering tensors in the Γ_{24} phase at the points T_1 and T_2 to be equal to the scattering tensors of the phases Γ_2 and Γ_4, respectively. We note that the RSL tensor of the softening magnon mode in the phases Γ_2 and Γ_4 never contains contributions of the magneto-optical constants of the given magnetically ordered phase.

Since the distance $\Delta\nu$ between two exchange branches is small in orthoferrites, $\Delta\nu \sim \nu^+ H_A H_E^{-1}$, in scattering by exchange branches the lines corresponding to different branches can merge into one if the spectrometer has insufficient frequency resolution. In this case the scattering tensor for the merged lines will be a superposition of the tensors of two exchange branches. This circumstance can be useful in an experimental search for exchange branches.

The analysis presented shows that the intensity of light scattering by exchange magnons should as a rule be less than the intensity of the scattering by the acoustic ones. This is possibly why no exchange branches of the spectrum were observed in Ref. 17, in an investigation of light scattering by the four-sublattice $NaNiF_3$, which orthoferrite structure and a magnetically ordered phase Γ_4, notwithstanding the undertaken special searches.

To conclude, it must be noted that light scattering by magnons in orthoferrites was recently investigated experimentally and theoretically in Ref. 18. The form of the RSL tensor was considered in there using the copresentations of the magnetic group.[1] The actual calculation, however, was based on an expansion of the dielectric constant in powers of **F** and **G** up to quadratic terms inclusive. As a result, the structure of the RSL tensor obtained in Ref. 18 coincides with our earlier expressions.[19] On the other hand, the exchange branches of the SW spectrum and light scattering by them were not considered in Ref. 18.

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APPENDIX

Frequencies of SW at $k = 0$ in magnetically ordered phase Γ_{24} in dimensionless units.

The acoustic branches:

$$\nu_{\mathrm{I}}^2 = E_1 K_4 \sin^2\theta \cos^2\theta,$$

$$\nu_{\mathrm{II}}^2 = E_1' \left\{ \left[E_{3yz} + D_{2y}^2 E_{1xz}^{-1} - D_{1z}^2 (E_4')^{-1} - (4S)^2 \left(e_3 \sin^2\theta + \frac{1}{2} e_2 \cos^2\theta \right) \right] \times \sin^2\theta + \left[E_{3yx} + D_{4y}^2 E_{1zx}^{-1} - D_{1z}^2 E_2^{-1} - (4S)^2 \left(e_1 \cos^2\theta + \frac{1}{2} e_2 \sin^2\theta \right) \right] \cos^2\theta \right\}.$$

The exchange branches:

$$\nu_{\mathrm{III}}^2 = E_4' E_2 + E_1' \left(D_{1z}^2 (E_4')^{-1} \sin^2\theta + D_{1x}^2 (E_2')^{-1} \cos^2\theta \right),$$

$$\nu_{\mathrm{IV}}^2 = E_4 E_2' + E_1 \left(D_{4z}^2 E_4^{-1} \sin^2\theta + D_{1x}^2 (E_2')^{-1} \cos^2\theta \right).$$

For the equations the following designations are used:

$$E_\alpha = E_{\alpha yx} \cos^2\theta + E_{\alpha yz} \sin^2\theta, \quad \alpha = 1, 2, 4,$$

$$E_\alpha' = E_{\alpha zx} \cos^2\theta + E_{\alpha xz} \sin^2\theta, \quad K_4 = e_2 - e_1 - e_3.$$

[1)]Allowance for the magnetoelastic interaction will leave the coefficients t_A, t_C, and d_G finite at the phase-transition points, but since the magnetoelastic interaction is weak in the vicinities of T_1 and T_2, these coefficients have a sharp maximum.

[2)]To simplify the expressions, we use in Eqs. (22)–(24) the notation $\lambda_1 = a_{xy,x}$, $\lambda_2 = a_{xy,y}$, $\lambda_3 = a_{xy,z}$ and $\lambda_5^{(i)} = a_{iixx}$, $\lambda_6^{(i)} = a_{iiyy}$, etc.

[3)]It can be easily seen that in both cases the direct products of the IR of the operators that participate in one mode contain IR of an equilibrium magnetic configuration.

[4)]Similar relations between the vectors of the weak and antiferromagnetism, magnetization, and antiferromagnetism are apparently valid for a large class of orthoferrites.

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