Antiferroelectric resonance in noncentrosymmetric multi-sublattice magnets

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(Submitted 19 February 1988)
Zh. Eksp. Teor. Fiz. **94**, 268–276 (September 1988)

We predict the phenomenon of antiferroelectric resonance (AFER), in which an AC electric field causes magnetic ions located at noncentrosymmetric positions in a multi-sublattice magnet to undergo magnetic transitions corresponding to exchange collective excitations of the system. We construct a theory of such resonances, and show that in magnets with collinear magnetic structures AFER is caused by relativistic and exchange-relativistic magnetoelectric interactions, while in the noncollinear magnets a significant contribution can also come from the exchange magnetoelectric interaction. We predict an exchange enhancement of the resonance by exchange modes, and discuss the role of AFER in causing resonant enhancement of magnetooptical phenomena. The main results of our theory are illustrated with the four-sublattice rhombohedral antiferromagnets α-Fe_2O_3 and Cr_2O_3 as examples.

I. INTRODUCTION

The tranditional method of investigating the resonance properties of magnetically-ordered crystals consists of subjecting them to a high-frequency (HF) magnetic field. Using this method we can reliably identify both acoustic modes (AM) and, in special cases, exchange modes (EM) of the crystal,[1–6] Excitation of EM with an HF magnetic field is possible only if the former are magnetically-active (i.e., coupled to oscillations of the ferromagnetism vector **M** of the system). However, the coupling of the EM to **M** comes about through relativistic and exchange-relativistic interactions, and therefore the intensity of absorption by EM exchange is weakened compared to AM exchange.[5–7] For nonmagnetically-active EMs interaction with a magnetic field is impossible, by virtue of selection rules (for example, in the case of EM which are odd under inversion).

In this paper we show that a natural way to resonantly excite magnetic-system exchange modes which are odd under inversion is to act on them with a HF electric field.[8] We construct a theory which describes the excitation of such modes by an AC electric field. The corresponding resonance will be referred to as antiferroelectric (AFER), because the EM are oscillations of the antiferromagnetism vectors $\mathbf{L}_i$. It is significant that in AFER the HF electric field is directly connected with the vectors $\mathbf{L}_i$ (our discussion centers on transitions directly induced by the AC electric field between the magnetic levels of the system).

The general condition for the existence of AFER will be formulated below. Here we only remark that in the majority of cases a sufficient condition for the existence of AFER in noncentrosymmetric crystals is the presence of magnetic ions at the noncentrosymmetric positions. Examples of such magnets are hematite, iron garnets, ferrite spinels, orthoferrites, and other compounds.

We will illustrate the essential features of AFER in noncentrosymmetric crystals with the examples of α-Fe_2O_3 and Cr_2O_3, where the former (according to Turov[9]) is even under inversion, while the latter is odd.

II. ANTIFERROMAGNETIC RESONANCE IN HEMATITE

As an example of a noncentrosymmetric crystal with a magnetic structure which is even under inversion, let us discuss the four-sublattice rhombohedral antiferromagnetic α-Fe_2O_3 (hematite). Following Ref. 10, we introduce the following linear combinations of the sublattice magnetizations $\mathbf{M}_i$ ($i = 1,2,3,4$):

$$\begin{aligned}\mathbf{M}&=\mathbf{M}_1+\mathbf{M}_2+\mathbf{M}_3+\mathbf{M}_4=4M_0\mathbf{m},\\ \mathbf{L}_1&=\mathbf{M}_1-\mathbf{M}_2-\mathbf{M}_3+\mathbf{M}_4=4M_0\mathbf{l}_1,\\ \mathbf{L}_2&=\mathbf{M}_1-\mathbf{M}_2+\mathbf{M}_3-\mathbf{M}_4=4M_0\mathbf{l}_2,\\ \mathbf{L}_3&=\mathbf{M}_1+\mathbf{M}_2-\mathbf{M}_3-\mathbf{M}_4=4M_0\mathbf{l}_3,\end{aligned} \tag{1}$$

where M_0 is the magnitude of the sublattice magnetization. In Table I we show the classification of the vectors (1) relative to the irreducible representations of the group D_{3d}^6 (for character tables of these irreducible representations and their notation see Ref. 11). The vectors **m** and $\mathbf{l}_1$, generate even, and $\mathbf{l}_2$ and $\mathbf{l}_3$ odd, representations of this group.

The system Hamiltonian has the form[10]

TABLE I. Magnetic configurations which are irreducible relative to the crystallographic group D_{3d}^6.

Irreducible representation	Irreducible spin configuration	Polarization of HF electric and magnetic fields	Irreducible representation	Irreducible spin configuration	Polarization of HF electric and magnetic fields
A_{1g}	l_{1z}	–	A_{1u}	l_{3z}	–
A_{2g}	m_z	h_z	A_{2u}	l_{2z}	E_z
E_g	(m^+, m^-), $(l_1^+, -l_1^-)$	(h^+, h^-)	E_u	(l_2^+, l_{-2}), $(l_3^+, -l_3^-)$	(E^+, E^-)

 0038-5646/88/091886-05$04.00

TABLE II. Classification of the homogeneous magnetic-resonance frequencies in α-Fe_2O_3 by symmetry type.

Ground State	Type of uniform oscillation	Variables	Components of $\chi_{ij}(\omega)$ which have a pole	Components of $\alpha_{ij}(\omega)$ which have a pole
A_{1g}, LO-phase	AM	m^+, l_1^+	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	–
	AM	m^-, l_1^-	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	–
	EM	l_2^+, l_3^+	–	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	l_2^-, l_3^-	–	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
E_g, LP-phase	AM	m_y, m_z, l_{1x}	$\chi_{yy}, \chi_{yz}, \chi_{zz}$	–
	AM	m_x, l_{1y}, l_{1z}	χ_{xx}	–
	EM	l_{2x}, l_{3y}, l_{3z}	–	α_{xx}
	EM	l_{2y}, l_{2z}, l_{3x}	–	$\alpha_{yy}, \alpha_{yz}, \alpha_{zz}$

$$(4M_0)^{-1}W=-{}^1/_2H_{e1}\mathbf{l}_1^2+{}^1/_2H_{e2}\mathbf{l}_2^2+{}^1/_2H_{e3}\mathbf{l}_3^2+{}^1/_2H_{e0}\mathbf{m}^2 +{}^1/_2H_{A1}l_{1z}^2+{}^1/_2H_{A2}l_{2z}^2+{}^1/_2H_{A3}l_{3z}^2+{}^1/_2H_{A0}m_z^2 +H_D(m_yl_{1x}-m_xl_{1y})+H_D'(l_{2x}l_{3y}-l_{2y}l_{3x})-(\mathbf{H}+\mathbf{h})\mathbf{m}. \quad (2)$$

Here H_{ei} is the effective field of the exchange interaction, H_{Ai} is the anisotropy field, and H_D, and H'_D are the Dzyaloshinskiĭ fields.

In the approximation linear in the electric field $\mathbf{E}(t)$ we must add to (2) the invariants (see Table I)

$$(4M_0)^{-1}\mathbf{EP}=E_x\{R_3(l_{1z}l_{3y}-l_{1y}l_{3z})+R_2(l_{1z}l_{2x}-l_{1x}l_{2z}) +r_3(m_xl_{3z}-m_zl_{3x})\}+E_y\{R_3(l_{1x}l_{3z}-l_{1z}l_{3x}) R_2(l_{1z}l_{2y}-l_{1y}l_{2z})+r_3(m_yl_{3z}-m_zl_{3y})\}+E_z\{\Pi_1\mathbf{m}\mathbf{l}_3+\Pi_2\mathbf{l}_1\mathbf{l}_2 +R_{3z}(l_{1y}l_{3x}-l_{1x}l_{3y})+r_{2z}(m_xl_{2y}-l_{2x}m_y)\}. \quad (3)$$

The antiferroelectric constants Π_i have their origin in exchange, while R_i and r_i are due to relativistic-exchange effects. We neglect terms of relativistic origin.

The dynamic properties of the system are described by the equations[1,2] $i\dot{\mathbf{m}}=[\mathbf{m}, W]$ and $i\dot{\mathbf{l}}_\nu=[\mathbf{l}_\nu, W]$, $\nu=1,2,3$. Linearizing these equations, we find that the AM correspond to oscillations of the vectors $\mathbf{m}$ and $\mathbf{l}_1$ which are even under inversion; these oscillations couple to the external magnetic field, i.e., they are magnetic dipole-active. The EM correspond to oscillations of the vectors $\mathbf{l}_2$ and $\mathbf{l}_3$, which do not interact with an external magnetic field or with the AM because they are odd under inversion. At the same time, an electric field couples to the vectors $\mathbf{l}_2$ and $\mathbf{l}_3$ (see Table I), i.e., the EM are electric dipole-active. These properties of the AM and EM are valid both for the easy-axis and easy-plane phases of hematite (see Table II).

Let us begin with an investigation of the EM in the easy-plane phase with a constant magnetic field $\mathbf{H}\|x$. Solving the linearized equations of motion, we find that a HF electric field $\mathbf{E}(t)\sim\mathbf{E}\exp(i\omega t)$ induces oscillations of the vectors l_2, $l_3\sim\exp(i\omega t)$ with amplitudes

$$l_{3z}=-\gamma^2R_3(H_{e1}+H_{e2})(\omega^2-\omega_{01}^2)^{-1}E_x, \quad (4)$$

$$l_{2z}=\gamma^2\{-R_2(H_{e1}+H_{e3})E_y+i[R_{3z} +(\Pi_2-\Pi_1)m]\omega E_z\}(\omega^2-\omega_{02}^2)^{-1}, \quad (5)$$

$$l_{3x}=\gamma^2\{[R_{3z}+(\Pi_2-\Pi_1)m](H_{e1}+H_{e2})E_z+i\omega R_zE_y\}(\omega^2-\varkappa_{02}^2)^{-1}. \quad (6)$$

$$l_{2y}=-\gamma^2R_2i\omega mE_y(\omega^2-\omega_{02}^2)^{-1}. \quad (7)$$

In this expression we include only those terms of exchange-relativistic origin which do not contain m; γ is the gyromagnetic ratio, and the EM frequency and magnetization are equal to

$$\gamma^{-2}\omega_{01}^2=(H_{e1}+H_{e3})(H_{e1}+H_{e2}) -[(H_{e0}-H_{e3})m+H_D'-H-H_D]H_{e1}m, \quad (8)$$

$$\gamma^{-2}\omega_{02}^2=(H_{e1}+H_{e3})(H_{e1}+H_{e2}+H_{A2}) +(H+H_D+H_D')[H+H_D+(H_{e2}-H_{e0})m],, \quad (9)$$

$$m=m_x=(H+H_D)(H_{e1}+H_{e0})^{-1}.$$

We remark that the spectrum of magnetic excitation of 4-sublattice hematite was calculated in Refs. 12–14; the results derived there coincide with Eqs. (8) and (9) [see also (17) below]. Inelastic neutron scattering experiments[12] show that EM in hematite are located in the infrared band.

To linear approximation in the spin deviations, we have from (3) for the components of the electric polarization vector:

$$P_x=-4M_0(R_3-r_3m)l_{3z}, \quad P_y=4M_0R_2l_{2z},$$
$$P_z=4M_0[(\Pi_2+r_{2z}m)l_{2y}+(-R_{3z}+\Pi_1m)l_{3x}].$$

Introducing the electric polarization tensor $\mathbf{P}(\omega)=\hat{\alpha}(\omega)\mathbf{E}(\omega)$, we find from Eqs. (4)–(7) and the relations above that the spin part of the HF electric polarization tensor $\hat{\alpha}(\omega)$ in the easy-plane phase with the magnetic field $\mathbf{H}\|x$ has the following nonzero components:

$$\alpha_{xx}(\omega)=4M_0\gamma^2R_3^2(H_{e1}+H_{e2})(\omega^2-\omega_{01}^2)^{-1}, \quad (10)$$

$$\alpha_{yy}(\omega)=4M_0\gamma^2R_2^2(H_{e1}+H_{e3})(\omega^2-\omega_{02}^2)^{-1}, \quad (11)$$

$$\alpha_{yz}(\omega)=i4M_0\gamma^2R_2[R_{3z}+(\Pi_2-\Pi_1)m]\omega(\omega^2-\omega_{02}^2)^{-1}, \quad (12)$$

$$\alpha_{zz}(\omega)=4M_0\gamma^2[R_{3z}+(\Pi_2-\Pi_1)m] \times[(-R_{3z}+\Pi_1m)(H_{e1}+H_{e2})](\omega^2-\omega_{02}^2]^{-1}. \quad (13)$$

In Eq. (10) we neglect the term r_3m compared to R_3, while in Eq. (13) we neglect $r_{2z}m$ compared to Π_2.

For the easy-axis phase ($l_{1z}=1$, $\mathbf{H}\|z$), we have

$$P_x=4(R_3l_{3y}+R_2l_{2x})M_0,$$
$$P_y=4R_2l_{2y}M_0, \quad P_z=\Pi_2l_{2z}4M_0. \quad (14)$$

Calculating the oscillation amplitudes of the antiferromagnetic vectors $\mathbf{l}_2(t)$ and $\mathbf{l}_3(t)$ under the action of AC electric field $\mathbf{E}(t)\sim\mathbf{E}\exp(i\omega t)$ in this phase and substituting them into Eqs. (14), we obtain the following nonzero spin contributions to the electric polarization tensor:

$$\alpha_{xx}(\omega)=\alpha_{yy}(\omega)=4M_0\gamma^2\{R_3^2(H_{e1}+H_{e2})+$$
$$+R_2^2(H_{e2}+H_{e3})\}(\omega_{01}\omega_{02}-\omega^2)(\omega_{01}^2-\omega^2)^{-1}(\omega_{02}^2-\omega^2)^{-1}, \quad (15)$$

$$\alpha_{xy}(\omega)=-\alpha_{yx}(\omega)=i4M_0\gamma^2\{R_3^2(H_{e1}+H_{e2})+$$
$$+R_2^2(H_{e2}+H_{e3})\}2\omega H(\omega_{01}^2-\omega^2)^{-1}(\omega_{02}^2-\omega^2)^{-1}. \quad (16)$$

Now, the EM frequency equals

$$\gamma^{-1}\omega_{01,2}=\{(H_{e1}+H_{e3}+|H_{A1}|)$$
$$\times(H_{e1}+H_{e2}+|H_{A1}|)-H_D'^2\}^{1/2}\pm H. \quad (17)$$

It is clear from Eqs. (10)–(13), (15) and (16) that in the easy-plane and easy-axis phases the residues at the EM poles of the HF electric polarization tensor are enhanced by exchange. The absorption intensity of the electric field at these frequencies is determined by the magnitude of the antiferroelectric constant; in what follows we give a numerical estimate of this constant.

Hematite is a straightforward example of a multi-sublattice system in which excitation of EM by a magnetic field is impossible by virtue of general selection rules, while for an AC electric field these rules allow such excitations. Let us now turn to a different system—Cr_2O_3.

III. ELECTRIC-DIPOLE-ACTIVE VIBRATIONS IN Cr_2O_3

The crystal Cr_2O_3 possesses a magnetic structure which is odd under inversion.[10] In the magnetic class which includes Cr_2O_3, inversion combines with the time-reversal operation $\bar{1}\cdot R$, and this results in a linear magnetoelectric effect.[16–18] The ground state is A_{1u}, with $\mathbf{l}_3\|z$. The magnetic properties of Cr_2O_3 are described by the potentials (2) and (3), in which it is necessary to make the replacements $H_{e1}\to -H_{e1}$, $H_{e3}\to -H_{e3}$, and $H_{A3}\to -H_{A3}$.

In the exchange approximation the acoustic type of oscillations in Cr_2O_3 correspond to transverse oscillations of the vectors $\mathbf{l}_3$ and $\mathbf{m}$, while those of exchange type correspond to transverse oscillation of the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$. Allowance for the Dzyaloshinksi interaction of H_D and H'_D in (2) leads to dynamic coupling of the EM and AM. Without pausing for detailed calculations we will present the final results.

A. Acoustic modes. Accurate to terms of order H_D/H_e and $(H_A/H_e)^{1/2}$ inclusively, the frequencies of the AM are equal to

$$\gamma^{-1}\omega_{A1,2}=\varepsilon_A\pm H, \quad (18)$$
$$\varepsilon_A^2=(H_{e3}+H_{e0})\{H_{A3}-H_D'^2(H_{e2}+H_{e3})^{-1})\}.$$

The nonzero transverse components of the HF magnetic susceptibility tensor have in this frequency interval the form

$$\chi_{xx}(\omega)=\chi_{yy}(\omega)=4M_0\gamma^2\{H_{A3}-H_D'^2(H_{e2}+H_{e3})^{-1}\}$$
$$\times(\omega^2-\varepsilon_A^2+H^2)(\omega^2-\omega_{A1}^2)^{-1}(\omega^2-\omega_{A2}^2)^{-1}, \quad (19)$$

$$\chi_{xy}(\omega)=-\chi_{yx}(\omega)=4iM_0\gamma^2\{H_{A3}-H_D'^2(H_{e2}+H_{e3})^{-1}\}$$
$$\times\omega H(\omega^2-\omega_{A1}^2)^{-1}(\omega^2-\omega_{A2}^2)^{-1}. \quad (20)$$

To linear order we have from (3) the electric polarization

$$P_x=4M_0(-R_3l_{1y}+r_3m_x),\quad P_y=4M_0(R_3l_{1x}+r_3m_y).$$

The nonzero spin contribution to the HF electric polarization tensor is described by the expressions

$$\alpha_{xx}(\omega)=\alpha_{yy}(\omega)=4M_0\gamma^2r_3^2\{H_{A3}-H_D'^2(H_{e2}+H_{e3})^{-1}\}$$
$$\times(\omega^2-\varepsilon_A^2+H^2)(\omega^2-\omega_{A1}^2)^{-1}(\omega^2-\omega_{A2}^2)^{-1}, \quad (21)$$

$$\alpha_{xy}(\omega)=-\alpha_{yx}(\omega)=i4M_0\gamma^2r_3^2\{H_{A3}-H_D'^2(H_{e2}+H_{e3})^{-1}\}$$
$$\times\omega H(\omega^2-\omega_{A1}^2)^{-1}(\omega^2-\omega_{A2}^2)^{-1}. \quad (22)$$

B. Exchange modes. To the same accuracy as above we have for the EM frequencies

$$\gamma^{-1}\omega_{01,2}=\varepsilon_0\pm H,$$
$$\varepsilon_0^2=(H_{e1}+H_{e3})(H_{e2}+H_{e3})+(H_{e1}+H_{e2}+2H_{e3})H_{A3}$$
$$+H_D'(H_D'+2H_D)(H_{e0}+H_{e3})(H_{e2}+H_{e3})^{-1}. \quad (23)$$

In this frequency interval,

$$\chi_{xx}(\omega)=\chi_{yy}(\omega)=-4M_0\gamma^2H_D'^2(H_{e2}+H_{e3})^{-1}\cdot$$
$$\times(\omega^2-\varepsilon_0^2+H^2)(\omega^2-\omega_{01}^2)^{-1}(\omega^2-\omega_{02}^2)^{-1}, \quad (24)$$

$$\chi_{xy}(\omega)=-\chi_{yx}(\omega)=4iM_0\gamma^2H_D'^2(H_{e2}+H_{e3})^{-1}$$
$$\times\omega H(\omega^2-\omega_{01}^2)^{-1}(\omega^2-\omega_{02}^2)^{-1}, \quad (25)$$

$$\alpha_{xx}(\omega)=\alpha_{yy}(\omega)=4M_0\gamma^2R_3^2(H_{e2}+H_{e3})$$
$$\times(\omega^2-\varepsilon_0^2+H^2)(\omega^2-\omega_{01}^2)^{-1}(\omega^2-\omega_{02}^2)^{-1}, \quad (26)$$

$$\alpha_{xy}(\omega)=-\alpha_{yx}(\omega)=-4iM_0\gamma^2R_3^2(H_{e2}+H_{e3})$$
$$\times\omega H(\omega^2-\omega_{01}^2)^{-1}(\omega^2-\omega_{02}^2)^{-1}. \quad (27)$$

Since the vectors ($\mathbf{l}_1$, $\mathbf{m}$), which are even under inversion, and the vectors ($\mathbf{l}_2$, $\mathbf{l}_3$), which are odd under inversion, particles in the EM and AM oscillations, these EM and AM can be excited both by magnetic and electric fields with $\mathbf{E}$, $\mathbf{h}\perp z$ (see Table III). However, from an experimental point of view the important thing is the magnitude of the absorption by the EM and AM. We obtain from (19)–(22) and (24)–(27) the following estimate of the susceptibility near the resonance frequency:

for EM,

$$\alpha(\omega_0)\sim\gamma M_0R^2(\omega-\omega_0)^{-1},\quad \chi(\omega_0)\sim\gamma M_0(H_A/H_e)(\omega-\omega_0)^{-1}.$$

for AM,

$$\alpha(\omega_A)\sim\gamma M_0R^2(H_A/H_e)^{1/2}(\omega-\omega_A)^{-1},$$
$$\chi(\omega_A)\sim\gamma M_0(H_A/H_e)^{1/2}(\omega-\omega_A)^{-1}.$$

The intensity of the absorption of an electric field by the EM is $(H_e/H_A)^{1/2}$ times larger than absorption by AM. For a

TABLE III. Classification of homogeneous magnetic resonance frequencies in Cr_2O_3 by symmetry type.

Ground State	Type of uniform oscillation	Variables (exchange approximation)	Components of $\chi_{ij}(\omega)$ which have a pole	Components of $\alpha_{ij}(\omega)$ which have a pole
A_{1u}, LO-phase	AM	m^+, l_3^+	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	AM	m^-, l_3^-	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	l_1^+, l_2^+	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$
	EM	l_1^-, l_2^-	$\chi_{xx}, \chi_{xy}, \chi_{yy}$	$\alpha_{xx}, \alpha_{xy}, \alpha_{yy}$

magnetic field, the situation is reversed: the intensity of the absorption by AM is $(H_e/H_A)^{1/2}$ times larger than that due to the EM. It is also clear that for $R^2 > H_A/H_e$, exitation of EM by an electric field is easier than by a magnetic field.

IV. PHYSICAL MECHANISM OF AFER

The physical mechanism which gives rise to the spin Hamiltonian is the same as the one discussed previously in constructing a theory of the magnetoelectric effect and of electric effects in paramagnetic resonance, and a theory of absorption and scattering of light in magnetically-ordered crystals.

A direct indication of the possibility of experimental observation of AFER is provided by experiments in which an AC electric field induces transitions between magnetic levels of paramagnetic ions in noncentrosymmetric sites, i.e., transitions from a state (l,m) to states $(l,m \pm 1)$ where l is the orbital and m the magnetic quantum number. Such transitions were observed in Refs. 19–20 (see also Ref. 22 and citations therein). As we have shown here, in centrosymmetric crystals which have high concentrations of magnetic ions and magnetic structures which are even under inversion (e.g., of Fe_2O_3 type) one consequence of these transitions will be magnetic excitations of exchange type.

The physics of AFER combines the physical mechanisms of electric-dipole paramagnetic resonance associated with impurity magnetic ions in noncentrosymmetric positions[21–24] and of absorption and scattering of light in systems with a high concentration of magnetic ions.[25,26,38] In systems with magnetic ions occupying centers of inversion, the electric dipole activity of the magnetic modes can be due to, e.g., the additional effect of a constant electric field.[27] We emphasize that in previously-studied magnets[28–33] without centers of inversion the effects investigated were due to coupling of electric and magnetic subsystems; in contrast, according to the theory constructed in this paper, AFER in noncentrosymmetric crystals is caused by direct excitation of exchange-type magnetic oscillations by an AC electric field.

We shall determine the antiferroelectric constants for α-Fe_2O_3 from experiments on the shift of the paramagnetic-resonance lines for Fe^{3+} ions.[22,34] These latter give an estimate of 10^{-2} for the single-ion spin-Hamiltonian constant. In systems with a high concentration of magnetic ions, contributions to the magnetoelectric effects come also from ion–ion interactions (in particular from exchange and relativistic-exchange interactions), which in individual cases increase the value of the constant by an order of magnitude.[35,36] Therefore in α-Fe_2O_3 (apparently) $R \sim 10^{-1}$. The value of II is also an order of magnitude larger.

It should also be noted that the contribution of invariants of exchange origin is proportional to the magnetization [see Eqs. (12) and (13)]; therefore, in the canted phases their contribution can significantly exceed the contribution from the exchange-relativistic invariants.

For quantitative estimates of the antiferroelectric interaction constants in Cr_2O_3 we make use of the results of experimental and theoretical studies of the magnetoelectric effect in this compound.[18,35,36] The latter give for the parameters of the spin Hamiltonian the values[35,36]

$$II \approx 5 \cdot 10^{-4}, \quad R \approx 5 \cdot 10^{-2}.$$

Taking into account that $H_A/H_e \sim 10^{-4}$ in Cr_2O_3, the conditions for observation of EM based on the absorption of an electric field can be more favorable than those based on absorption of a magnetic field.

V. LINEAR RESONANT MAGNETOELECTRIC EFFECTS

Let us formulate the general conditions for electric-dipole activity of the magnetic oscillations.

In the general case the EM and AM are of electric-dipole type if the dynamic compounds of the antiferromagnetism and ferromagnetism vectors which correspond to them transform according to the irreducible representations of the unitary subgroup of the system's magnetic symmetry group (i.e., they transform just like the compounds of the electric polarization vector **P**).

Let us investigate in more detail the general conditions for the existence of AFER in magnetic structures whose ground state in the exchange approximation is collinear and satisfies the conditions of the Turov classification[9] (see also Ref. 39).

In centrosymmtric crystals it is necessary to distinguish between structures which are even and odd under inversion.

1. The structures $\bar{1}(+)$. If a system whose structure is even under inversion contains magnetic ions which are not located at inversion centers, then in addition to the basic even antiferromagnetism vector $\mathbf{L}_0$ there exists at least one other antiferromagnetism vector **l** which is odd under inversion. In the thermodynamic potential of such a magnet it is possible to have invariants of the form

$$K_{\alpha\beta\gamma} E_\alpha L_{0\beta} l_\gamma, \quad \alpha, \beta, \gamma = x, y, z. \tag{28}$$

In a static electric field, Eq. (28) leads to the appearance of $l_\gamma \sim E_\alpha$. In analogy with the magnetoelectric effect this phenomenon can be called antiferroelectric (AFEE).

In an AC electric field $\mathbf{E} \sim \exp(i\omega t)$, the relation (28) causes oscillation of the vector **l**, and for $\omega = \omega_0$ exctiation of EM (ω_0 is the exchange frequency).

2. For structures $\bar{1}(-)$ odd under inversion, it is necessary to distinguish between two cases according to parity relative to a translation **t**.

Systems with $\bar{1}(-)$ and $\mathbf{t}(+)$ pertain to antiferromagnets whose thermodynamic potentials include an invariant of the form

$$K_{\alpha\beta\gamma} L_{0\alpha} M_\beta E_\gamma, \tag{29}$$

where **M** is the magnetic moment of the system. If in this case there also exists an antiferromagnetic vector **l** which is odd under inversion, then it is also possible to have an invariant of the form (28). This invariant gives rise to the presence of AFER and AFEE, while (29) makes possible excitation of AM by an electric field, i.e., magnetoelectric resonance.

In systems with $\bar{1}(-)$ and $\mathbf{t}(-)$ (the magnetic unit cell is larger than the crystallographic unit cell) the invariant (29) is forbidden and there is no static magnetoelectric effect. However, an invariant of the form (28) is possible, where now the antiferromagnetic vector **l** is odd under translation and even under inversion. In this system there will be both AFEE and AFER.

IV. CONCLUSION

We shall dwell in somewhat greater detail on the possibility of experimentally observing antiferroelectric resonance with EM and AM.

Apparently, low-dimensionality magnets are most convenient from an experimental point of view for observing EM by resonant methods. In such systems, because of the presence of weak exchange interactions, the EM and AM frequencies are comparable as a rule,[3,6] Examples of low-dimensionality magnet in which it is possible to observe AFER, are the eight-sublattice antiferromagnets $CsMnCl_2 \cdot 2H_2O$ and $CuCl_2 \cdot C_4H_8SO$. We note that EM were recently observed in the AFMR spectrum of $CuCl_2 \cdot C_4H_8SO$ (see Ref. 40). As our analysis shows, in this compound the EM can also be observed in the AFER spectrum by placing the sample at an electric field antinode within a cavity. The AM can be observed by using the same method.

The EM frequencies of α-Fe_2O_3 and Cr_2O_3 are located in the infrared wavelength band. Therefore the features of experimental observation of AFER in these compounds are close to those in optical experiments.[26] If the dimensions of the sample are comparable with or smaller than the wavelength of the AC electric field, then the theory developed above is directly applicable.

For bulk samples of α-Fe_2O_3 and Cr_2O_3, the equations of motion for the vectors (1) must be considered jointly with the Maxwell equations. In this case, the exchange spin modes and the electromagnetic oscillations are found to be coupled, which leads to their mutual restructuring. A detailed analysis of this question is outside the framework of the present communication; therefore we will only pause briefly to treat a specific case.

For $\mathbf{E} \perp \mathbf{k} \| z$ the dispersion relation or coupled right-hand-polarized electromagnetic waves and EM ω_{01} (17) in hematite has the form

$$\left[\left(\frac{kc}{n_+}\right)^2 - \omega^2\right](\omega_{01}-\omega)(\omega_{02}+\omega) = \frac{4\pi}{n_+^2}\alpha'\omega^2. \qquad (30)$$

Here c is the velocity of light while the parameter α' which determines the value of the coupling of the branches is

$$\alpha' = 4M_0\gamma^2\{R_3^2(H_{e1}+H_{e2}) + R_2^2(H_{e2}+H_{e3})\},$$

n_+ is the refractive index of the medium without allowance for the EM contribution. The dispersion of the EM in this frequency band is not significant and we disregard it. For left-hand-polarized waves the dispersion relation is obtained from (30) by the replacements $\omega_{01} \leftrightarrow \omega_{02}$, $n_+ \leftrightarrow n_-$.

As is clear from (19)–(22) and (24)–(27), when magnetic oscillations can be excited by an electromagnetic field it is possible to have resonance singularities both in the components of the magnetic susceptibility tensor and in the components of the electric polarization tensor. It is necessary to take this circumstance into account both in development of a theory of propagation of electromagnetic waves and in analysis of the experimental data. In particular, near the exchange resonances it is possible to resonantly enhance such magnetooptic effects as Faraday rotation, the Cotton-Mouton effect, and the Kerr effect.

It should also be kept in mind that, in addition to the linear processes we have investigated here, the Hamiltonian (3) also contributes to two-magnon absorption, to parametric instability, and to other nonlinear phenomena, even in an approximation linear in the electric field.

The authors wish to express their gratitude to V. G. Bar'yakhtar, V. V. Eremenko, A. I. Zvyagin, and A. A. Stepanov for discussions of this work.

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Translated by F. Crowne