Acoustics of magnetoelectric antiferromagnetics: tetragonal crystals

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The acoustic properties and characteristics of antiferromagnetic resonance (AFMR) which are associated with the magnetoelectric effect are studied for tetragonal antiferromagnetics which have a center of antisymmetry and exhibit the magnetoelectric effect. In conjunction with the preceding work [E. A. Turov, Zh. Eksp. Teor. Fiz.], which is devoted to rhombohedral antiferromagnetics, this makes it possible to discuss how the most interesting such effects vary with the type of magnetic structure and magnetic state and as a function of the direction of the magnetic and electric field vectors relative to the crystallographic axes.

INTRODUCTION

The acoustic properties associated with the magnetoelectric effect (which occurs in centrally antisymmetric magnetic structures) in rhombohedral antiferromagnetics were studied in Ref. 1. Antiferromagnetics with the structure $1^-3_z^+2_x^-$, typical of Cr_2O_3, were studied. Unfortunately, the latter compound is an easy-axis antiferromagnetic (EAA), and the expected effects (dependence of the sound speed on the electric field, acoustic birefringence of magnetoelectric nature, and others) are stronger for easy-plane antiferromagnetics (EPA). We do not know of any specific rhombohedral EPA, but such antiferromagnetics do exist among tetragonal crystals. Examples are the trirutiles Cr_2TeO_6 (T_N=105 K), V_2WO_6 (T_N=370 K), and others.[2–5] Another example[4] could be $FeSb_2O_4$ (T_N=46 K).

Our main problem is to establish how magnetoelectric effects influence the acoustic properties as a function of the type of magnetic structure and magnetic state of the antiferromagnetic and as a function of the magnitude and direction of the magnetic field (H) and electric field (E). Tetragonal antiferromagnetics are also interesting in that after magnetic ordering occurs the four-fold crystal-chemical symmetry axis in these materials can become either even ($4\rightarrow4^+$) or odd ($4\rightarrow4^-$). As will be seen below, for these two cases the effects of interest to us can be qualitatively different.

Just as in Ref. 1, we consider only low frequencies

$$\omega \ll \omega_{\mathrm{AFMR}}, \tag{1}$$

where ω_{AFMR} is the AFMR frequency. This condition makes it possible to find the renormalization of the elastic moduli $C_{\alpha\beta\gamma\delta}$ due to magnetoelastic and magnetoelectric interactions using the fact that for these frequencies the vectors **M** and **L** of the magnetic subsystem, and especially the electric polarization vector. **P**, follow in a quasiequilibrium manner the elastic deformations $e_{\alpha\beta}$, without solving the equations of magnetoelastic dynamics. We note, however, that the AFMR frequencies are then also determined at the same time, since they enter, in a known manner, into the renormalized moduli $C_{\alpha\beta\gamma\delta}$. This was especially necessary to do, since, as far as we know, these frequencies have not been calculated for tetragonal magnetoelectric antiferromagnetics (some results for the rhombohedral case are contained in the paper by V. G. Shavrov[6]).

In the concluding section the results of this paper for tetragonal antiferromagnetics are discussed and compared with the results for rhombohedral case (Ref. 1). We would like not only to separate the most interesting magnetoelectric effects in acoustics and in magnetic resonance but also to give some recommendations, based on specific quantitative estimates, for materials and experiments; conditions. Unfortunately, in many cases the lack of experimental data on the parameters appearing in the theoretical formulas makes this problem very difficult. But, at the same time, it is precisely experiments of this type that provide additional opportunities for determining the indicated parameters.

The calculations are performed using a model in which the sublattice magnetizations have constant moduli

$$M_1^2=M_2^2=M_0^2, \tag{2}$$

where the magnetic susceptibility $\gamma_\parallel$ parallel to the antiferromagnetism vector $L=2M_0l$ is zero, $\chi_\parallel=0$. In order to clarify the role of nonzero susceptibility ($\chi_\parallel\neq0$), however, it is desirable to perform calculations using a different model in which (**ML**) and therefore $\chi_\parallel$ are different from zero (see, for example, Ref. 7). But this will be done elsewhere.

1. THERMODYNAMIC POTENTIAL

We consider tetragonal antiferromagnetics with centrally antisymmetric magnetic structures of the form $\bar{1}^-4_z^+2_d^-\equiv\bar{1}^-4_z^+2_{??}^{??}$ and $\bar{1}^-4_z^-2_d^\mp\equiv\bar{1}^-4_z^-2_x^\pm$. Such structures occur in a number of trirutiles[2–5] as well as rare-earth phosphates and vanadates—compounds of the type $HoPO_4$ and $GdVO_4$[8–10] (see also the citations in Ref. 4). Of these compounds, terbium phosphate $TbPO_4$ (T_N=2.28 K) has the highest (among known magnetoelectric antiferromagnetics) magnetoelectric susceptibility $\alpha=P/H=M/E$ $=1.1\cdot10^{-2}$ (in the CGS system).[10]

The total density of the thermodynamic potential

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$$F=F_M+F_P+F_e+F_{le}+F_{MP}$$

consists of the magnetic (F_M), electric polarization (F_P), elastic (F_d), (antiferro)magnetic elastic (F_{le}), and magnetoelectric contributions. With the exception of F_{MP}, all other energies have the same form for both types of structures (with 4_z^+ and 4_z^-):

$$F_M=\tfrac{1}{2}\chi^{-1}\mathbf{M}^2-(\mathbf{MH})+\tfrac{1}{2}K(l_x^2+l_y^2)\,(\text{or }\tfrac{1}{2}Kl_z^2)+\tfrac{1}{2}K_2l_x^2l_y^2, \tag{3}$$

$$F_P=\tfrac{1}{2}\varkappa_\perp^{-1}(P_x^2+P_y^2)+\tfrac{1}{2}\varkappa_z^{-1}P_z^2-(\mathbf{PE}), \tag{4}$$

$$F_e=\tfrac{1}{2}C_{11}(e_{xx}^2+e_{yy}^2)+C_{12}e_{xx}e_{yy}+C_{13}(e_{xx}+e_{yy})e_{zz}+\tfrac{1}{2}C_{33}e_{zz}^2 +2C_{66}e_{xy}^2+2C_{44}(e_{xz}^2+e_{yz}^2), \tag{5}$$

$$F_{le}=B_{11}(l_x^2e_{xx}+l_y^2e_{yy})+B_{12}(l_x^2e_{yy}+l_y^2e_{xx})+2B_{66}l_xl_ye_{xy} +2B_{44}(l_xe_{xz}+l_ye_{yz})l_z. \tag{6}$$

At the same time the magnetoelectric contributions have a substantially different form, depending on the parity of the 4_z axis. Thus for $4_z^+2_d^-$ and $4_z^-2_d^-$ we have, respectively:[6]

$$(4_z^+)F_{MP}=-\gamma_2(l_xP_x+l_yP_y)M_z-\gamma_3(M_xP_x+M_yP_y)l_z -\gamma_4(l_xM_x+l_yM_y)P_z-\gamma_5l_zM_zP_z, \tag{7}$$

$$(4_z^-)F_{MP}=-\gamma_2(l_xP_y+l_yP_x)M_z-\gamma_3(M_xP_y+M_yP_x)l_z -\gamma_4(M_xl_y+M_yl_x)P_z. \tag{8}$$

We now make several remarks concerning the expressions written above for the energies (3)–(8).

1) In contrast to Ref. 1, the anisotropy in the basal plane (the constant K_2) is taken into account in the magnetic energy. We assume also that K_2 contains a contribution owing to spontaneous deformations.

2) The expression F_P [Eq. (4)] is identical to that from (1.16), F_{MP} [Eq. (7)] is a particular case of (1.15) with $\gamma_1=0$, and F_{le} [Eq. (6)] is a particular case of (1.17) with $B_{14}=B_{41}=0$.

3) The expression for F_{MP} (8) is written for the structure $1^-4_z^-2_d^-$, which, as already mentioned, occurs in some trirutiles. However, a simple transformation, namely, a rotation by 45° around the z axis (so that the 2_d and 2_x axes are interchanged, $2_d\rightleftarrows 2_x$), transforms the expression into a form suitable for describing the structure $\bar{1}^-4_z^-2_d^+\equiv\bar{1}^-4_z^-2_x^-$, characteristic for some of the above-mentioned rare-earth phosphates. Then we obtain for F_{MP} instead of Eq. (8)

$$F_{MP}=-\gamma_2(l_xP_x-l_yP_y)M_z-\gamma_3(M_xP_x-M_yP_y)l_z -\gamma_4(l_xM_x-l_yM_y)P_z. \tag{9}$$

4) Of course, the energy F_{MP} of Eq. (8) for the structure $\bar{1}^-4_z^-2_d^-$ will have the form (9) if the 2_d^- axis is taken as the new x' axis. We note that Eq. (9) differs from Eq. (7) in that in all parentheses the "+" sign is replaced by a "−" and γ_5 is set equal to zero.

The foregoing remarks will help us sometimes to use the results for a single structure in order to obtain results for a different structure without performing new calculations.

Just as in Ref. 1, the calculations are performed in the following order. First, by minimizing $\tilde{F}_{MP}=F_M+F_P+F_{MP}$ with respect to M and P, we put F_{MP} into the form

$$\tilde{F}_{MP}=\tfrac{1}{2}\tilde{K}_{\alpha\beta}l_\alpha l_\beta, \tag{10}$$

in which the effective "anisotropy constants" $\tilde{K}_{\alpha\beta}$ determine, in particular, the AFMR frequencies. Next, by minimizing $\tilde{F}_{MP}+F_{le}$ we find the dynamical values $l_\alpha=f(e_{\beta\gamma})$ induced by the acoustic deformations e_δ. After substituting them into $\tilde{F}_{MP}$ and F_{le} [Eq. (10) + Eq. (6)] we obtain the correction to F_e (5), described by the renormalized moduli:

$$\tilde{C}_{\alpha\beta\gamma\delta}=C_{\alpha\beta\gamma\delta}+\Delta C_{\alpha\beta\gamma\delta}(\mathbf{H},\mathbf{E}).$$

We now present the final results, first for the structures with 4_z^+ and then for structures with 4_z^-. Here we confine our attention only to effects which are linear in the parameters $\gamma_i\sim\alpha$, if they exist.

2. THE STRUCTURE $\bar{1}^-4_z^+2_d^-(2_x^-)$

As already mentioned above, for this structure the results can be obtained from the formulas given in Ref. 1, setting $\gamma_1=B_{14}=B_{41}=0$ and adding the basal-anisotropy constant K_2 to K_x in the state $\mathbf{L}\parallel y$. Correspondingly, for the states $\mathbf{L}\parallel z$(A) and $\mathbf{L}\parallel y\parallel z$(B) we obtain

A. EASY-AXIS ANTIFERROMAGNETIC, $\mathbf{L}\parallel z$

A1. $\mathbf{H}\parallel\mathbf{E}\parallel\mathbf{L}\parallel z$

$$\tilde{F}_{MP}=\tfrac{1}{2}(K_xl_x^2+K_yl_y^2), \tag{11}$$

where

$$K_x=K_y=K-\chi(H_z-\gamma_0l_z^0\varkappa_zE_z)^2\equiv\tilde{K}. \tag{12}$$

Here $l_z^0=\pm1$, depending on the direction of the vector $\mathbf{l}$ in the domains. For otherwise the same conditions $\gamma_0l_z^0>0$ ($\gamma_0\equiv\gamma_4-\gamma_5$) corresponds to minimum ground-state energy. Next we find

$$l_x=-\frac{2B_{44}l_z^0}{\tilde{K}}e_{xz},\quad l_y=-\frac{2B_{44}l_z^0}{\tilde{K}}e_{yz}. \tag{13}$$

As a result, the elastic constant C_{44} is renormalized so that

$$\tilde{C}_{44}=\tilde{C}_{55}=C_{44}-B_{44}^2/\tilde{K}. \tag{14}$$

The transverse acoustic modes with wave vector $\mathbf{k}\parallel\mathbf{Z}$ are then softened, but they remain degenerate:

$$(\Omega_{\mathbf{k}}^2)_{1,2}=(\tilde{C}_{44}/\rho)k_z^2\equiv v_\perp^2k_z^2. \tag{15}$$

The condition for magnetoelastic stability of the state under consideration with $\mathbf{H}\parallel\mathbf{E}\parallel\mathbf{L}\parallel z$ has the form $\tilde{C}_{44}\geqslant0$ or

$$\tilde{K}\geqslant B_{44}^2/C_{44}. \tag{16}$$

With some experience in calculating spin-wave and magnetoelastic spectra, knowing the quantity $\tilde{K}$ defined in Eq. (12), it is possible to write down expressions for the AFMR frequencies. This is most difficult to do in this case of axial symmetry, since the dynamical variables l_x and l_y do not belong to different sets of normal coordinates (the cyclic variables $l_{\pm}=l_x \pm i l_y$ are such coordinates; see, for example, Ref. 11). For this reason, as already indicated in Ref. 1, the "anisotropy constant" $\tilde{K}$ defined in Eq. (12) determines only the product of two AFMR frequencies $\omega_1\omega_2/\gamma^2=\tilde{K}\chi^{-1}$ (γ is the gyromagnetic ratio). Knowing the limiting values of ω_1 and ω_2 for $E=0$,[11] however, they can be written out separately for $E\neq 0$ also:

$$\begin{aligned} \omega_1&=\gamma\sqrt{2H_EH_A}-(H_z-\gamma_0 l_z^0 \varkappa_z E_z), \\ \omega_2&=\gamma\sqrt{2H_EH_A}+(H_z-\gamma_0 l_z^0 \varkappa_z E_z). \end{aligned} \tag{17}$$

Here $H_E=M_0/\chi$ and $H_A=K/2M_0$ are the effective homogeneous exchange field and the uniaxial magnetic anisotropy field.

A2. H|| E|| *x* [see Eqs. (29)–(31) in Ref. 1]

In this case the different constants K_x and K_y are obtained with the previous form of $\tilde{F}_{MP}$, [given in Eq. (11)]:

$$K_x=K+\chi H_x[H_x+(2\gamma_2+3\gamma_3)l_z^0\varkappa_\perp E_x], \tag{18'}$$

$$K_y=K+\chi\varkappa_\perp \gamma_3 l_z^0 H_x E_x. \tag{18''}$$

Here

$$l_x=-\frac{2B_{44}l_z^0}{K_x}e_{xz}, \quad l_y=-\frac{2B_{44}l_z^0}{K_y}e_{yz}, \tag{19}$$

which, in contrast to Eq. (14), leads to different renormalization of the elastic moduli:

$$\tilde{C}_{44}=C_{44}-B_{44}^2/K_y, \ \tilde{C}_{55}=C_{44}-B_{44}^2/K_x. \tag{20}$$

This means that the degeneracy of the transverse acoustic modes with $k\parallel z$ is removed and linear acoustic birefringence (BR) is obtained:

$$\begin{aligned} \frac{\Delta v}{v}&\equiv\frac{v_{\perp x}-v_{\perp y}}{v_\perp}=\frac{\tilde{C}_{55}-\tilde{C}_{44}}{2C_{44}}\approx\frac{B_{44}^2}{2C_{44}K}(K_x-K_y) \\ &\approx\left(\frac{B_{44}}{K}\right)^2\frac{\int H_x[H_x+2\varkappa_\perp(\gamma_2+\gamma_3)l_z^0E_x]}{2C_{44}}. \end{aligned} \tag{21}$$

(The subscripts x and y for $v_\perp$ indicate the direction of polarization of the corresponding wave.)

Thus the magnetoelectric effect causes $\Delta v/v$ to acquire a dependence on the field E, the sign of the magnetoelectric correction being determined by the sign of the product $(\gamma_2+\gamma_3)l_z^0$ for the corresponding domain.

Turning next to the AFMR frequencies, we note that l_x and l_y in Eq. (11) now refer to different collections of dynamical variables, corresponding to normal vibrations. This leads to the relations

$$\omega_1^2/\gamma^2=\chi^{-1}K_x, \quad \omega_2^2/\gamma^2=\chi^{-1}K_y. \tag{22}$$

For $E=0$ the well-known results in the approximation H_x, $H_A \ll H_E$ are obtained (except for Ref. 11; see also Ref. 12).

B. EASY-PLANE ANTIFERROMAGNETIC, L|| *y*

B1. H|| E|| *x* [see Ref. 1, Eqs. (39)–(45)]

Here

$$\tilde{F}_{MP}=\frac{1}{2}(K_x l_x^2+K_z l_z^2), \tag{23}$$

where

$$K_x=K_2+\chi H_x^2-\gamma_2^2\chi(\varkappa_\perp E_x)^2,$$

$$K_y=K-\gamma_3^2\chi\varkappa_\perp(\chi H_x^2+\varkappa_\perp E_x^2).$$

The magnetoelectric effect, linear in γ_i, does not occur in this case, but the magnetoelectric contribution to K_x "competes" only with the small basal anisotropy K_2. Correspondingly, K_x determines the lower AFMR frequency

$$\omega_1^2/\gamma^2=K_x\chi^{-1}, \tag{24}$$

and K_z determines the upper AFMR frequency

$$\omega_2^2/\gamma^2=K_z\chi^{-1}. \tag{25}$$

Acoustic oscillations l are now determined by the formulas

$$l_x=-\frac{2B_{66}l_y^0}{K_x}e_{xy}, \quad l_z=-\frac{2B_{44}l_y^0}{K_z}e_{yz} \quad (l_y^0=\pm 1) \tag{26}$$

which leads to renormalization of the two elastic moduli

$$\tilde{C}_{66}=C_{44}-B_{66}^2/K_x; \quad \tilde{C}_{44}=C_{44}-B_{44}^2/K_z; \quad \tilde{C}_{55}=C_{44}. \tag{27}$$

Here the magnetoelectric interaction also contributes to birefringence, but this effect is stronger for other directions of **H** and **E**, and we shall study it in greater detail for this other geometry, when **H**|| z and **E**|| y.

B2. H|| *z*, E|| *y*(L|| *y*)

This situation was not considered in Ref. 1. This made it necessary to perform a special calculation, the results of which will also be applicable for the structure $\bar{1}^-3_z^+2_x^-$ (if $K_2=0$). These results (in the approximation linear in γ_i) are as follows.

The effective anisotropy (23), the AFMR frequencies (24) and (25), l_x and l_z (26), as well as the renormalized moduli $\tilde{C}_{66}$ and $\tilde{C}_{44}$ (27) have the same form as in the preceding case. The constants K_x and K_z are, however, entirely different:

$$K_x=K_2+\gamma_2 l_y^0\chi\varkappa_\perp H_z E_y, \tag{28}$$

$$K_z=K+\chi H_z^2+(\gamma_2+2\gamma_3)l_y^0\chi\varkappa_\perp H_z E_y. \tag{29}$$

The quantity $l_y^0=\pm 1$ appearing in these formulas takes into account the possible existence of 180°-degree domains. It should be kept in mind, however, that for tetragonal EPA there can exist, in general, both 180° and 90° domains. In order to eliminate the latter it is sufficient to perform annealing in a magnetic field. For example, in the case $H\parallel X$ there will remain domains with $l\uparrow\uparrow Y$ and $l\uparrow\downarrow Y$, which, strictly speaking, the formulas (28) and (29) presuppose. If, however, annealing is then performed in crossed fields $H \| Z$ and $E \| Y$, then due to the magnetoelectric interaction [the term with γ_2 in the energy (7)] the energetically most favorable state should be established. This state corresponds to the condition

$$\gamma_2 l_y^0 > 0. \tag{30}$$

But, the last annealing by itself is, in principle, sufficient in order to obtain this state. The absolute criterion for the state under consideration with $L \| Y$, $H \| Z$, and $E \| Y$ to be stable [taking into account the condition (30)] is that the squared frequencies of the acoustic modes with $k \| Y$, which interact dynamically with the M and P subsystems, must be non-negative. These will be

$$\Omega_{ik}^2 = (\tilde{C}_{66}/\rho) K_y^2, \quad \Omega_{2k}^2 = (\tilde{C}_{44}/\rho) k_y^2, \tag{31}$$

where $\tilde{C}_{66}$ and $\tilde{C}_{44}$ are determined by the expressions (27) using Eqs. (28) and (29).

The indicated requirement determines the minimum possible value of the effective anisotropy constants:

$$K_x \gg \frac{B_{66}^2}{C_{66}} \quad \text{and} \quad K_z \gg \frac{B_{44}^2}{C_{44}}. \tag{32}$$

The equality corresponds to phase-transition points. The latter can be approached "from above," if the condition "greater than" in the relations (32) is achieved due to sufficiently strong fields H and E, after which these fields are decreased. These limiting minimum values of K_x and K_z at the phase-transition points correspond to minimum values of the AFMR frequencies (24,25), determining the so-called magnetoelastic gap for magnons [we recall that K_x and K_y are taken, in this case, from Eqs. (28) and (29)]:

$$\begin{aligned} \omega_{1\,\min} &\equiv \omega_{me1} = \gamma \sqrt{2H_E H_{me1}}, \\ \omega_{2\,\min} &\equiv \omega_{me2} = \gamma \sqrt{2H_E H_{me2}}, \end{aligned} \tag{33}$$

where

$$H_{me1} = B_{66}^2/2C_{66}M_0, \quad H_{me2} = B_{44}^2/2C_{44}M_0 \tag{34}$$

are the effective magnetoelastic fields.[13]

The magnetoelastic acoustic modes (31) must become substantially softer near phase transitions. From the standpoint of the magnetoelectric effect, in this respect, the first mode is most interesting. In accordance with Eq. (32) the condition for this mode to be stable can be rewritten in the form

$$|\gamma_2| H_z(\varkappa_\perp E_y) \gg 2H_E(H_{me1} - H_\Box), \tag{35}$$

where

$$H_\Box = K_2/2M_0$$

is the effective tetragonal magnetic anisotropy field. For $H_\Box > 0$ the equality in the relation (35) can hold only if $H_{MY1} > H_\Box$. However, the condition (35) can also be realized for $H_\Box < 0$. In both cases the fundamental possibility arises here that due to the magnetoelectric effect the acoustic mode will be softened in crossed fields H_z and E_y. Of course, the magnetoelectric effect can also be observed directly according to the shift in the lower AFMR frequency:

$$\omega_1 = \gamma [2H_E H_\Box + |\gamma_2| \varkappa_\perp H_z E_y]^{1/2}. \tag{36}$$

We now consider the acoustic birefringence, using Eq. (27) with Eqs. (28) and (29). For waves with $k \| z$, polarized along the x and y axes, we have

$$\frac{\Delta v}{v} = \frac{\tilde{C}_{55} - \tilde{C}_{44}}{2C_{44}} = \frac{B_{44}^2}{2C_{44}K_z}. \tag{37}$$

For waves with $k \| y$, polarized along the x and z axes [the frequencies of the waves are determined by Eqs. (31)] we obtain

$$\frac{\Delta v}{v} = \frac{\tilde{C}_{44} - \tilde{C}_{66}}{2C_{44}} \simeq \frac{C_{44} - C_{66}}{2C_{44}} + \frac{B_{66}^2}{2C_{44}K_x}. \tag{38}$$

In the latter equality we have taken into account the fact that $K_x \ll K_z$.

In the first case (37) birefringence is entirely associated with the magnetoelastic interaction, and the influence of the magnetoelectric interaction is weak. In the second case birefringence already exists due to the purely crystallographic anisotropy of the elastic properties, but here the antiferromagnetic correction to $\Delta v/v$ can be quite noticeable due to the small value of K_x (28). One can hope that this is the most favorable situation for observing the effect of the magnetoelectric interaction on the acoustic birefringence.

3. THE STRUCTURE $\bar{1}^- 4_z^- 2_d^- (2_x^+)$

We consider once again easy-axis and easy-plane antiferromagnetics with this structure for different directions of the fields **E** and **H**, giving preference to cases when magnetoelectric effects linear in γ_i are present.

C. EASY-AXIS ANTIFERROMAGNETIC, L$\|$ z.

C1. H$\|$ E$\|$ z.

Here it is convenient to use the coordinate system (x', y', z), rotated with respect to the x and y axes [in which the magnetoelectric interaction (a) is written] by 45° around the z axis. In these coordinates the magnetoelectric energy assumes the form (9) with x and y replaced by x' and y', all other energies (3)–(6) having their previous form (though it should be kept in mind that the constants C_{ij} and B_{kl}, being linear combinations of the old constants, will be new).

This makes it possible to write down immediately, using the results of the preceding Sec. A_1, an answer for the present case. The point is that Eqs. (9) and (7) for F_{MP} differ only through the signs in the parentheses (in addition, in our last case $\gamma_5 = 0$). Therefore the corresponding results can be obtained from Sec. A_1 by simply changing the sign of γ_0 ($\equiv \gamma_4$) in K_y (12) as compared to K_x, so that we obtain from Eq. (12)

$$\begin{aligned} K_x &= K - \chi (H_z - \gamma_4 l_z^0 \varkappa_z E_z)^2, \\ K_y &= K - \chi (H_z + \gamma_4 l_z^0 \varkappa_z E_z)^2, \end{aligned} \tag{39}$$

and Eqs. (13) and (14) must be replaced by

$$l_x=-\frac{2B_{44}l_z^0}{K_x}e_{xz},\quad l_y=-\frac{2B_{44}l_z^0}{K_y}e_{yz};\tag{40}$$

$$\tilde{C}_{55}=C_{44}-\frac{B_{44}^2}{K_x},\quad \tilde{C}_{44}=C_{44}-\frac{B_{44}^2}{K_y}.\tag{41}$$

In order to determine ω_1 and ω_2 we have once again only one relation $\omega_1^2\omega_2^2/\gamma^4=K_xK_y\chi^{-2}$ (in this case the tip of the vector $\mathbf{l}$ describes ellipses around $\mathbf{H}$). Knowing the value of ω_1 and ω_2 in the limit $E=0$ we find

$$\begin{aligned}\omega_1^2/\gamma^2&=[\sqrt{K\chi^{-1}}-(H_z-\gamma_4l_z^0\varkappa_zE_z)]\\&\times[\sqrt{K\chi^{-1}}-(H_z+\gamma_4l_z^0\varkappa_zE_z)],\\\omega_2^2/\gamma^2&=[\sqrt{K\chi^{-1}}+(H_z-\gamma_4l_z^0\varkappa_zE_z)]\\&\times[\sqrt{K\chi^{-1}}+(H_z+\gamma_4l_z^0\varkappa_zE_z)].\end{aligned}\tag{42}$$

Writing this differently

$$\omega_{1,2}^2/\gamma^2=[(\sqrt{K\chi^{-1}}\mp H_z)^2-(\gamma_4\varkappa_zE_z)^2]\tag{43}$$

we can see that, in contrast to Eq. (17), here the contribution of the magnetoelectric interaction to the AFMR frequencies is quadratic in $\gamma_4E\sim\alpha E$.

In accordance with Eqs. (41) and (39), however, birefringence will be linear in γ_4 and will be due entirely to the magnetoelectric interaction:

$$\frac{\Delta v}{v}=\frac{\tilde{C}_{55}-\tilde{C}_{44}}{2C_{44}}=\left(\frac{B_{44}}{K}\right)^2\frac{\gamma_4l_z^0\chi\varkappa_zH_zE_z}{C_{44}},\tag{44}$$

where $\Delta v/v$ has opposite signs for AFM domains with $l_z^0=+1$ and $l_z^0=-1$. It is in principle possible to "see" domains acoustically.

C2. H|| E|| x'|| 2_d^-

In this situation the results are described by the same formulas as in the case A_2, so that only one of the two terms (for example, $M_{x'}P_{x'}-M_{y'}P_{y'}$), namely, the first one, is significant. The final formulas are obtained from Eqs. (18)–(22) by setting $\gamma_5=0$ (i.e., $\gamma_0\to\gamma_4$).

If H|| E|| y', then, conversely, the first term becomes important. Thus only the sign change $\gamma_4\to-\gamma_4$ need be made in the indicated formulas. It should be kept in mind, however, that the equilibrium state will now correspond to the condition $\gamma_4l_z^0<0$.

C3. H|| x|| 2_x^+, E|| y

This is another relative arrangement of $\mathbf{H}$ and $\mathbf{E}$ (recall that we have assumed $\mathbf{L}\|z$) for which a linear magnetoelectric effect exists. Here we have returned to the initial coordinate system, and the expression (8) must be used for $\tilde{F}_{MP}$. The results are as follows: $\tilde{F}_{MP}$ has the form (11), where

$$K_x=K+\chi H_x^2+(2\gamma_2+3\gamma_3)l_z^0\chi\varkappa_\perp H_xE_y,\tag{45'}$$

$$K_y=K+\gamma_3l_z^0\chi\varkappa_\perp H_xE_y.\tag{45''}$$

We present also the results for birefringence and the AFMR frequencies:

$$\frac{\Delta v}{v}=\frac{\tilde{C}_{55}-\tilde{C}_{44}}{2C_{44}}=\left(\frac{B_{44}}{K}\right)^2\frac{\chi H_x[H_x+2(\gamma_2+\gamma_3)l_z^0\varkappa_\perp E_y]}{2C_{44}};\tag{46}$$

$$\omega_1^2/\gamma^2=K_x\chi^{-1},\quad \omega_2^2/\gamma^2=K_y\chi^{-1}.\tag{47}$$

Here, birefringence is probably once again the most interesting magnetoelectric effect.

D. EASY-PLANE ANTIFERROMAGNETIC, L⊥ z

D1. H|| x, E|| z, L|| y

$\tilde{F}_{MP}$ is determined by Eq. (23) with

$$K_x=K_2+\chi H_x^2+5\gamma_4l_y^0\chi\varkappa_zH_xE_z,\tag{48'}$$

$$K_z=K+\gamma_4l_y^0\chi\varkappa_zH_xE_z.\tag{48''}$$

With these values of K_x and K_z the results for $\omega_{1,2}$, $l_{x,z}$, $\tilde{C}_{66}$, and $\tilde{C}_{44}$ are determined, respectively, by Eqs. (24) and (25) and Eqs. (26) and (27), and birefringence is determined by Eqs. (37) and (38).

The lower AFMR frequency can be rewritten, using Eqs. (24) and (48), in a form similar to Eq. (36):

$$\omega_1=\gamma[2H_EH_\square+H_x(H_x+5|\gamma_4|\varkappa_zE_z)]^{1/2}.\tag{49}$$

Finally, we have one other case for which, compared to the case considered above, the fields H and E are interchanged.

D2. H|| z, E|| x, L|| y

The formulas for $\tilde{F}_{MP}$ (23), ω_1^2 (24) and ω_2^2 (25), l_x and l_z (26), and $\tilde{C}_{ij}$ (27) remain the same, if K_x and K_z are expressed as

$$K_x=K_2+\gamma_3\,l_y^0\chi\varkappa_\perp\,H_zE_x,\tag{50'}$$

$$K_z=K+\chi H_z^2+3\gamma_3\,l_y^0\chi\varkappa_\perp\,H_zE_x.\tag{50''}$$

The equations (37) and (38) for the birefringence retain their previous form for $K\|Z$ and $K\|Y$, respectively, and the lower AFMR frequency (24) becomes

$$\omega_1=\gamma[2H_EH_\square+|\gamma_3|\varkappa_\perp\,H_zE_x]^{1/2}.\tag{51}$$

This equation is similar to Eq. (36) for the structure $\bar{1}^-4_z^+2_d^-$ in the state L|| y, H|| z, and E|| y (see Sec. B_2), and all considerations regarding phase transitions and the role of the magnetoelastic gap are also valid here. In particular, the stability condition (35) in this case assumes the form

$$|\gamma_3|H_z(\varkappa_\perp E_x)\geqslant 2H_E(H_{me1}-H_\square).\tag{52}$$

4. THE STRUCTURE $\bar{1}^-4_z^-2_d^+(2_x^-)$

This magnetic structure occurs in the phosphates of rare-earth metals $HoPO_4$ (T_N=1.4 K), $DyPO_4$ (T_N> 3.5 K), $TbPO_4$ (2.13 K < T < T_N=2.28 K), and others (see Refs. 5, 8, and 9 and the citations there).

Their magnetoelectric interaction (9) is obtained from the magnetoelectric interaction for the structure $\bar{1}^-4_z^-2_d^-(2_x^+)$, i.e., from Eq. (8), by rotating the coordinates x and y around the z axis by 45°. For this reason, one can simply use the computational results from Sec. 3.

For the states $\mathbf{L}\parallel z$, in fields $\mathbf{H}\parallel \mathbf{E}\parallel z$ and $\mathbf{H}\parallel \mathbf{E}\parallel x$, this will be the formulas from Secs. C1 and C2, respectively, with x' and y' replaced by x and y. For fields $\mathbf{H}\parallel [110]$ and $\mathbf{E}\parallel [1\bar{1}0]$, in the same state, it is necessary to transform to a coordinate system where the indicated axes (the 2_d^+ symmetry axes) are used as, respectively, the x' and y' axes. Then F_{MP} (9) assumes the form (8). For this case the formulas from Sec. C3, with x and y replaced by x' and y' should be used.

The latter applies completely to the state $\mathbf{L}\parallel y$, for which the formulas of the Secs. D (D1 and D2) can be rewritten simply, once again, of course, keeping in mind the new axes $(x'\parallel 2_d^+)$.

We underscore once again that the choice of the directions of the fields $\mathbf{H}$ and $\mathbf{E}$ is not accidental. It is dictated by the consideration that linear magnetoelectric effects must be present.

CONCLUSIONS

This and the preceding paper (Ref. 1) actually concern the effect of an electric field, via the magnetoelectric interaction, on the acoustic properties and AFMR frequency of antiferromagnetics. As regards the AFMR frequency, besides the theoretical work cited[6] there is also Ref. 14, in which an electric-field-induced shift of the AFMR frequency was observed experimentally in rhombohedral chromium oxide, Cr_2O_3. An explanation is given on the basis of the assumption that the magnetic anisotropy constant and the g-factor depend on E. Similar ideas are elaborated in Ref. 15. We cannot describe here either the ideas or the experimental results. We merely note the fact that the indicated effect was observed experimentally in a situation that is far from the most favorable one. We do not know of any works concerning the influence of the magnetoelectric effect on the acoustics of antiferromagnetics.

The aim of our analysis of the role of the magnetoelectric interaction in acoustics (and the associated effects in AFMR) for different AFM structures and states in fields H and E with different directions was not only to study theoretically the corresponding magnetoelectric effects but also to determine the most favorable specific materials and geometric conditions for observing these effects experimentally. In addition, the existence or absence itself of any specific magnetoelectric effect can sometimes solve unequivocally the question of the choice of magnetic structure from two (or more) possible structures in compounds of the same type (for example, in trirutiles).

In this respect the AFM structures $\bar{1}^-4_z^+2_d^-$ and $\bar{1}^-4_z^-2_d^-$ (both structures are encountered in trirutiles) in the state $\mathbf{H}\parallel \mathbf{E}\parallel \mathbf{L}\parallel 4_z$ are typical. For the first of these structures acoustic birefringence is absent in this state, while for the second structure it should occur [compare Eqs. (14)–(15) and Eqs. (41), (44)]. Further, the AFMR frequency shift is linear in the magnetoelectric field αE for the first structure and quadratic for the second structure [compare Eqs. (17) and (43)].

Turning now to the most favorable situations for observing magnetoelectric effects (birefringence in acoustics and magnetoelectric frequency shift in AFMR), we note, first, that these two groups of phenomena are organically interrelated. This is understandable; after all both groups are determined by the same quantities, the "effective anisotropy constants" K_α which we introduced above [see, for example, Eqs. (24)–(25) and (37)–(38)]. These effects are strongest near orientational-phase-transition points, where the corresponding constants K_α are minimum.

We call attention to different situations in which the field αE can manifest itself in competition with other factors.

The first and most interesting situation is when αE competes only with the magnetic anisotropy and the field H only intensifies the magnetoelectric effect [see Eqs. (18″), (28), (44), (45″), (48″), and (50′)]. In this case the magnetoelectric effects should be strongest in easy-plane antiferromagnetics, in which the field αE "competes" only with the small basal anisotropy (K_2). This is situation B_2 [for example, Eqs. (35)–(36)] or situation D_2 [Eqs. (51)–(52)], as well as similar situations for the structure $\bar{1}^-4_z^-2_d^+$ ($\mathbf{L}\perp z$), Sec. 4. In this case the magnetoelectric field αE plays a very appreciable role, if $\alpha EH \gtrsim K_2$ holds, which is a completely realistic possibility. For example, for $\alpha=10^{-4}$ (larger values are also possible) and $K_2=10^2$ ergs/cm^3 for $H=10^4$ Oe we have $E \geqslant 10$ kV/cm.

In the second case the effective field αE must compete with the magnetic field H [see Eqs. (12), (17), and (39)]. But most often H, on the one hand, increases the absolute magnitude of the magnetoelectric effect, while on the other it masks this effect, since H and E appear as the combination $\chi H^2+\alpha HE$ [see Eqs. (18′), (21), (29), (45′), (46), (48′), (49), and (50″)].

We must still mention the possible situations for a magnetoelectric effect in acoustic birefringence. First, birefringence can arise entirely due to the magnetoelectric interaction. This should happen for easy-axis antiferromagnetics, when $\mathbf{K}\parallel \mathbf{H}\parallel \mathbf{E}\parallel \mathbf{L}\parallel 4_z$ for structures $\bar{1}^-4_z^-2_d^\mp$ [see, for example, Eq. (44)]. Setting in Eq. (44) $(B_{44}/K)^2 \approx 10^4$, $\alpha=10^{-2}$ [$TbPO_4$ (Ref. 8)], $\chi=10^{-3}$, $H=1$ T, and $E=10$ kV/cm we obtain for $\Delta v/v$ a value of several percent.

Another case also occurs for easy-axis antiferromagnetics with $\mathbf{k}\parallel z$, but for $\mathbf{H}\perp z$, when birefringence now exists due to the magnetic field, while αE merely intensifies the effect [see, for example, Eqs. (21) and (46)]. Finally, for $\mathbf{k}\perp 4_z$ birefringence now exists due to the crystallographic elastic anisotropy, and the magnetoelectric interaction (together with magnetoelastic interaction) makes only an additional contribution, which depends on H and E [see, for example, Eq. (38)]. An estimate made using the indicated formula shows that this effect could also be observable experimentally.

We did not find it possible here to make quantitative estimates of the magnetoelectric effects of interest to us for specific magnetoelectric antiferromagnetics. This requires a very thorough analysis of existing experimental data on the parameters appearing in the theory.

We wish to say a few words about omissions in our papers and future work. First, additional calculations must be performed using a different model of an antiferromagnetic (in which $(\mathbf{LM})\neq 0$ holds and therefore, the parallel magnetic susceptibility satisfies $\chi_{\parallel}\neq 0$), especially for the state $\mathbf{H}\parallel \mathbf{L}\parallel 4_z$ at higher temperatures. Second, of course, it is also desirable to study orthorhombic magnetoelectric antiferromagnetics (for example, orthoaluminates; see Ref. 2). Finally, the modulation obtained here of the vector $\mathbf{l}$ by sound, resulting in acoustic modulation of the AFM part of the dielectric permittivity, makes it possible to study diffraction of light by sound in such antiferromagnetics and the effect of the magnetoelectric interaction on such diffraction.

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