Acoustics of magneto-electric antiferromagnetics. Rhombohedral crystals

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We calculate the effective contribution of the magnetoelastic interaction to the elastic moduli for centrosymmetric antiferromagnetics in the region of low frequencies which are small compared to the antiferromagnetic-resonance frequency, where the magnetizations of the magnetic sublattices and the electric polarizability follow the acoustic deformation in quasiequilibrium. There is a linear magnetoelectric effect in those antiferromagnetics. As a result the elastic moduli, renormalized with these interactions taken into account, and hence also the sound velocity, turn out to depend not only on the magnetic field, as in antiferromagnetics with central symmetry, but also on the electric field. We consider rhombohedral antiferromagnetics, amongst them especially Cr_2O_3.

INTRODUCTION

The linear magnetoelectric effect occurs in crystals in which the symmetry allows the existence in the thermodynamic potential of terms of the form

$$F_{HE}=-\hat{\alpha}\mathbf{HE}, \tag{1}$$

where $\hat{\alpha}$ is the magnetoelectric susceptibility tensor and $\mathbf{H}$ and $\mathbf{E}$ are the magnetic and electric field-strength vectors. It is clear that this is possible only in systems without a symmetry center (space inversion). There is also no time inversion ($t\to -t$), as indicated by the existence of a magnetic structure.[1]

In antiferromagnetics there can occur a situation where a symmetry center $\bar{1}$ which exists in a crystal-chemical (Fedorov) symmetry group is transformed into an antisymmetry center $\bar{1}'$ after magnetic ordering. This means, in fact, that here the crystal-chemical inversion couples the magnetic atoms pertaining to magnetic sublattices with opposite magnetization directions. The vector parameter $\mathbf{L}$ of the antiferromagnetic ordering ($\mathbf{L}=\mathbf{M}_1-\mathbf{M}_2$ in a two-sublattice model) changes its sign under the action of $\bar{1}$ into its opposite:

$$\bar{1}\mathbf{L}=-\mathbf{L}. \tag{2}$$

We have assumed in Eq. (1) that there is invariance with respect to the elements of the true (magnetic) symmetry. In accordance with the basic crystal-physics rules (see, e.g., Ref. 2, Sec. 25) it is then sufficient to consider only the elements of the magnetic point symmetry for the actual magnetic structure and magnetic state under consideration. We take the magnetic structure to mean the relative directions of the magnetic moments with respect to one another, and the magnetic state to mean their direction (e.g., that of the vector $\mathbf{L}$) relative to the crystallographic axes. If we are interested in the properties of the antiferromagnetic under external actions (and with a change of temperature) under which the magnetic state (and, in general, the magnetic structure) and with it the magnetic symmetry change, we must start from the crystal-chemical symmetry in as far as it remains unchanged.

To do this it is necessary to take into account explicitly the dependence of $\hat{\alpha}$ in (1) on the antiferromagnetism vector $\mathbf{L}$ which disturbs the initial crystal-chemical symmetry. It usually turns out to be sufficient to expand $\hat{\alpha}$ in powers of $\mathbf{L}$ and to limit oneself to the linear terms. In other words, we shall write the magnetoelectric part of the thermodynamic potential density in the form:

$$F_{MP}=-\gamma_{\alpha\beta\gamma}l_\alpha M_\beta P_\gamma, \tag{3}$$

where $\mathbf{l}=\mathbf{L}/2M_0$ (M_0 is the actual modulus of the sublattice magnetization) while $\mathbf{M}$ and $\mathbf{P}$ are, respectively, the total (local) magnetization and the electric polarizability. In contrast to (1) we introduced here the internal variables $\mathbf{M}$ and $\mathbf{P}$ bearing in mind the non-equilibrium thermodynamic potential necessary for the dynamics.

Since we take out the vector $\mathbf{l}$, the "breaker" of the crystal-chemical symmetry, explicitly we must determine the actual form of the tensor $\hat{\gamma}$ from the requirements of crystal-chemical symmetry. It is then necessary to take into acount the parity of the symmetry elements.[3,4] We call elements g^+ and g^- even and odd, respectively, if they couple magnetic atoms pertaining to one and the same magnetic sublattice (and also to sublattices with the same magnetization direction) or to sublattices with opposite magnetizations. It is important to remember that if $g^{\pm}\mathbf{M}=g\mathbf{M}$ and $g^{\pm}\mathbf{P}=g\mathbf{P}$ (similarly for $\mathbf{H}$ and $\mathbf{E}$) we have $g^{\pm}\mathbf{L}=\pm g\mathbf{L}$, where g is a point-symmetry element acting on $\mathbf{M}$ and $\mathbf{L}$ as on normal axial vectors, and on $\mathbf{P}$ as on a polar vector (Eq. (1) is a special case of the expressions given here since the antisymmetry center directly corresponds to an odd inversion: $\bar{1}'\equiv\bar{1}$). Taking the parity of the elements into account reflects, indeed, the fact that we are here dealing already with elements of space (crystal chemical) symmetry.

In terms of the parity of the symmetry elements one can easily classify an antiferromagnetic (AFM) structure (we shall deal here with the case when the magnetic and

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the crystal chemical elementary cells are the same). The classification must include the independent symmetry elements (generators of the crystal chemical group) with an indication of their parity. For instance, the AFM structure of Cr_2O_3 (space group $R\bar{3}c$) can be classified as $\bar{1}\ 3_z^+ 2_x^-$. The threefold axis $3\|\ z$ is even whereas the inversion $\bar{1}$ and the twofold axis $2\|\ x$ are odd. To obtain from the relevant classification the corresponding AFM structure (when knowing the positions of the magnetic atoms) we must direct the magnetic moments of the atoms which are coupled by even symmetry elements parallel to one another and those which are coupled by odd symmetry elements antiparallel to one another (here $g_1^{\pm} g_2^{\pm} = g_3^+$ and $g_1^{\pm} g_2^{\mp} = g_3^-$).

We can obtain the explicit form of the thermodynamic potential, including the magnetoelectric part (3), from the requirement that it be invariant with respect to the symmetry elements which occur in the classification, taking their parity into account. We must also add to the magnetoelectric interaction energy (3) the magnetic (exchange, Zeeman, and magnetic anisotropy) energy

$$F_M = \frac{1}{2}\chi^{-1}\mathbf{M}^2 - \mathbf{MH} + \frac{1}{2}K_{\alpha\beta} l_\alpha l_\beta + \frac{1}{2}\mathscr{E}_{\beta\gamma}\frac{\partial l_\alpha}{\partial x_\beta}\frac{\partial l_\alpha}{\partial x_\gamma} + \ldots, \tag{4}$$

the magnetoelastic (or more precisely, the antiferromagnetic-elastic) energy

$$F_{le} = B_{\alpha\beta\gamma\delta} l_\alpha l_\beta e_{\gamma\delta}, \tag{5}$$

where

$$e_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha}\right)$$

is the deformation tensor ($\mathbf{u}$ is the elastic displacement vector); and, finally, the energy of the electric polarization $\mathbf{P}$:

$$F_P = \tfrac{1}{2}\varkappa_{\alpha\beta}^{-1} P_\alpha P_\beta - \mathbf{PE}, \tag{6}$$

where $\varkappa_{\alpha\beta}$ is the electric polarizability tensor.

Our problem consists in studying the effect of the magnetoelectric interaction (1) through $\mathbf{M}$ and *le* interactions [corresponding to (4) and (5)] on the acoustic properties of the antiferromagnetic. We shall consider the simplest (and, generally speaking, the most interesting) case of relatively low frequencies

$$\omega \ll \omega_{\mathrm{AFMR}}, \omega_P, \tag{7}$$

where ω_{AFMR} is the AFM resonance frequency ($\approx 10^{10}$–$10^{11}\ \mathrm{s}^{-1}$) which is the minimum frequency for spin waves while ω_P is the frequency of the polarization oscillations (optical frequency region). One can under those conditions assume that $\mathbf{M}$, $\mathbf{l}$, and $\mathbf{P}$ follow the dynamic elastic deformations $e_{\alpha\beta}$ in quasi-equilibrium, in agreement with the minimum of the thermodynamic potential

$$F = F_M + F_P + F_{MP} + F_{le}. \tag{8}$$

Moreover, one can for those frequencies neglect in F_M in (4) the inhomogeneous exchange (the term with $\mathscr{E}_{\beta\gamma}$).

The calculation reduces to that first of all we determine from the equations

$$\frac{\partial F}{\partial \mathbf{M}} = \frac{\partial F}{\partial \mathbf{P}} = \frac{\partial F}{\partial \mathbf{l}} = 0 \tag{9}$$

the dynamic quantities $\Delta\mathbf{M}$, $\Delta\mathbf{P}$, and $\Delta\mathbf{l}$ caused by the acoustic oscillations $e_{\alpha\beta}$. The values found for $\Delta\mathbf{M}$, $\Delta\mathbf{P}$, and $\Delta\mathbf{l}$ expressed in terms of the $e_{\alpha\beta}$ are later substituted into F in (8). The quadratic form in the $e_{\alpha\beta}$ thus obtained (if we restrict ourselves to the harmonic approximation) must be added to the elastic energy density

$$F_e = \tfrac{1}{2}C_{\alpha\beta\gamma\delta} e_{\alpha\beta} e_{\gamma\delta}, \tag{10}$$

and as a result we get a renormalization of the elastic moduli:

$$C_{\alpha\beta\gamma\delta} \to \tilde{C}_{\alpha\beta\gamma\delta} = C_{\alpha\beta\gamma\delta} + \Delta C_{\alpha\beta\gamma\delta}(\mathbf{H},\mathbf{E}). \tag{11}$$

These moduli then become dependent on $\mathbf{H}$ and $\mathbf{E}$, and acoustic birefringence and other effects appear which are connected with magneto-elasticity and magneto-electricity. From purely symmetry arguments (without energy considerations), some of them were predicted in a paper by the present author[5] (see also Ref. 4).

A few words about the other approximations (apart from the ones mentioned above) used in the calculations.

Firstly, we neglect that the longitudinal (for $\mathbf{H}\|\ \mathbf{l}$) susceptibility $\chi_\|$ of the antiferromagnetic is nonvanishing. This corresponds to a model in which

$$\mathbf{Ml} = 0 \tag{12}$$

and

$$\mathbf{L}^2 + \mathbf{M}^2 = \mathrm{const}$$

and which with more justification is applicable in the low-temperature region where $\chi_\| \ll \chi_\perp \equiv \chi$.

Secondly, we assume that the magnetoelectric susceptibility [compare (1) and (3)]

$$\alpha_{\mu\nu} = \gamma_{\alpha\beta\gamma} l_\alpha \chi_{\beta\mu} \varkappa_{\gamma\nu} \tag{13}$$

is a sufficiently small quantity although it may for different antiferromagnetics vary within very broad limits: $\alpha = 10^{-5}$–10^{-2} (Refs. 6–8). For instance, $\alpha_{\max} \approx 6 \cdot 10^{-4}$ for Cr_2O_3 ($T_N = 310$ K) while for $\alpha_{\max} \approx 1 \cdot 10^{-2}$ $TbPO_4$ ($T_N = 2.3$ K).[8] This makes it possible to restrict ourselves in all calculations of effects of interest to us to an approximation of not higher than second order in the parameter $\hat{\gamma} \sim \hat{\alpha}$.

We have already mentioned that in the present paper we shall base our considerations on the example of the $\bar{1}\ 3_z^+ 2_x^-$ structure. We shall consider here two magnetic states: the "easy axis" kind when $\mathbf{L}\|\ 3\|\ z$ and the "easy plane" kind when $\mathbf{L}\perp 3$ for various directions of the $\mathbf{H}$ and $\mathbf{E}$ fields.

1. THE L|| z STATE

We first write down general expressions for the energies contained in F in (8) which are invariant for the $\bar{1}\ 3_z^+ 2_x^-$ structure:[9]

$$F_M = \tfrac{1}{2}\chi^{-1}\mathbf{M}^2 - \mathbf{MH} + \tfrac{1}{2}K(l_x^2 + l_y^2), \tag{14}$$

$$F_{M\mathscr{P}}=-\gamma_1[(l_xM_y+l_yM_x)P_x+(l_xM_x-l_yM_y)P_y]$$
$$-\gamma_2(l_xP_x+l_yP_y)M_z-\gamma_3l_z(M_xP_x+M_yP_y)$$
$$-\gamma_4(l_xM_x+l_yM_y)P_z-\gamma_5l_zM_zP_z, \quad (15)$$

$$F_P=\tfrac{1}{2}\varkappa_\perp^{-1}(P_x^2+P_y^2)+\tfrac{1}{2}\varkappa_z^{-1}P_z^2-\mathbf{PE}, \quad (16)$$

$$F_{le}=B_{11}(l_x^2e_{xx}+l_y^2e_{yy})+B_{12}(l_x^2e_{yy}+l_y^2e_{xx})+B_{33}l_z^2e_{zz}$$
$$+2B_{66}l_xl_ye_{xy}+2B_{44}(l_yl_ze_{yz}+l_xl_ze_{xz})+2B_{14}[(l_x^2$$
$$-l_y^2)e_{yz}+2l_xl_ye_{xz}]+B_{41}[l_yl_z(e_{xx}-e_{yy})$$
$$+2l_xl_ze_{xy}]. \quad (17)$$

We note that when $K>0$ in (14) the anisotropy energy corresponds to an easy-axis antiferromagnet. For an easy-plane antiferromagnet it is more convenient to write it as $Kl_z^2/2$ (again we assume that $K>0$). We neglect the anisotropy in the base plane. It is usually small for trigonal symmetry. Furthermore, by virtue of the condition (12) we can eliminate in (15) one of the last two terms (with γ_4 and γ_5) introducing a new constant of the magnetoelectric interaction:

$$\gamma_0=\gamma_4-\gamma_5.$$

We consider two cases for the directions of the **H** and **E** fields:

1) $\mathbf{H}\parallel\mathbf{E}\parallel z$

Assuming that $P_z=\varkappa_zE+\Delta P_z$ we can choose as the independent variables in this case M_x, M_y, l_x, l_y, P_x, P_y, and ΔP_z. According to (12) we then have

$$M_z=-(M_xl_x+M_yl_y)/l, \quad (18)$$

$$l_z\approx l-(l_x^2+l_y^2)/2l, \quad (19)$$

where $l\approx\pm1$.

In those variables, and retaining in $\tilde{F}_{MP}\equiv F_M+F_P+F_{MP}$ [see (14)–(16)] only terms which are quadratic in them, we have

$$\tilde{F}_{MP}=\tfrac{1}{2}\chi^{-1}(M_x^2+M_y^2)+\tfrac{1}{2}K(l_x^2+l_y^2)+(M_xl_x+M_yl_y)$$
$$\times(H/l-\gamma_0\varkappa_zE)-\gamma_3l(M_xP_x+M_yP_y)$$
$$+\tfrac{1}{2}\varkappa_\perp^{-1}(P_x^2+P_y^2)+\tfrac{1}{2}\varkappa_z\Delta P_z^2. \quad (20)$$

Minimizing $\tilde{F}_{MP}$ with respect to **M** and **P** we find

$$\tilde{F}_{MP}=\tfrac{1}{2}\tilde{K}(l_x^2+l_y^2), \quad (21)$$

where

$$\tilde{K}=K-\chi_\perp\frac{(H-\gamma_0l\varkappa_zE)^2}{1-\gamma_3^2\chi\varkappa_\perp}. \quad (22)$$

Adding to $\tilde{F}_{MP}$ of (21) the remaining term F_{le} from (17) and then minimizing $F=\tilde{F}_{MP}+F_{le}$ with respect to l_x and l_y we get

$$l_x=-2l(B_{41}e_{xy}+B_{44}e_{xz})/\tilde{K},$$
$$l_y=-l[B_{41}(e_{xx}-e_{yy})+2B_{44}e_{yz}]/\tilde{K}. \quad (23)$$

The modulation of the antiferromagnetism vector **l** by the acoustic deformations $e_{\alpha\beta}$, which is described by Eqs. (23), must lead to a corresponding modulation of the antiferromagnetic (**l**-dependent) part of the permittivity.

After substituting the values of l_x and l_y from (23) into the energy $F=\tilde{F}_{MP}+F_{le}$ we find the required effective contribution to the elastic energy:

$$\tilde{F}_{MP}+F_{le}\to\Delta F_e$$
$$=-\tilde{K}^{-1}\{2B_{41}^2e_{xy}^2+2B_{44}^2(e_{xz}^2+e_{yz}^2)$$
$$+2B_{41}B_{44}[(e_{xx}-e_{yy})e_{yz}+2e_{xy}e_{xz}]\}. \quad (24)$$

Adding this to the initial elastic energy (10) of a rhombohedral crystal we see easily that its renormalized part has the form:

$$\tilde{F}_e=2\tilde{C}_{66}e_{xy}^2+2\tilde{C}_{44}(e_{xz}^2+e_{yz}^2)+2c_{41}[(e_{xx}-e_{yy})e_{yz}$$
$$+2e_{xy}e_{xz}], \quad (25)$$

where

$$\tilde{C}_{66}=C_{66}-B_{41}^2/\tilde{K},\quad \tilde{C}_{44}=C_{44}-B_{44}^2/\tilde{K}$$
$$\tilde{C}_{41}=C_{41}-B_{41}B_{44}/\tilde{K}.$$

There occurs thus a softening of the acoustic modes connected with these elastic moduli. The condition for the stability of the state considered with $\mathbf{l}\parallel z$ (for $\mathbf{H}\parallel\mathbf{E}\parallel z$) is then that all these moduli be positive-definite, i.e.,

$$\tilde{C}_{44}\geqslant0 \quad \text{for } \tilde{K}\geqslant B_{44}^2/C_{44} \quad (26)$$

and not the condition $\tilde{K}>0$ as might seem to follow from (21). In the phase transition ("spin flop") point there appears a magnetoelastic gap so that the value of $\tilde{K}$ of (22) which determines it remains finite and one of the acoustic modes is softened (see in this connection Ref. 10).

We call attention to the important fact that if the magnetoelectric interaction is taken into account $\tilde{K}$ of (22), and hence the energy of the whole system, depends on the sign of the product γ_0l. From the two states with $l=\pm1$ the more stable ground state corresponds to the one for which $\gamma_0l>0$. Such a choice will occur only when both fields are nonvanishing: $H\neq0$ and $E\neq0$. It is just this fact which makes it possible to remove the AMF domain structure in magnetoelectrics through heat treatment while simultaneously applying **E** and **H** (see, e.g., Ref. 6).

Incidentally, the quantity $\tilde{K}$ of (22) determines the gap for spin waves (the AMF frequencies ω_1 and ω_2 of Ref. 9). More precisely, in the present case $\tilde{K}\chi^{-1}=\omega_1\omega_2/\gamma^2$ (γ is the gyromagnetic ratio).

We consider next acoustic waves with wave vectors $\mathbf{k}\parallel\mathbf{z}$. According to (25) and (22) their velocity is determined by the expression

$$v^2=\frac{\tilde{C}_{44}}{\rho}\approx\frac{1}{\rho}\left[C_{44}-\frac{B_{44}^2}{K-\chi(H-\gamma_0l\varkappa_zE)^2}\right], \quad (27)$$

from which it is clear that this velocity will be different for domains with opposite directions of **l** ($l=\pm1$). This fact opens up the possibility in principle of "acousto-vision" of domains in magnetoelectric antiferromagnetics.

Of course, all these effects show up most strongly near the "spin-flop" point which corresponds to the equal sign in (26).

2) $\mathbf{E}\| \mathbf{H}\| \mathbf{x}$

In this case $M_x=\chi H+\Delta M_x$ and $P_x=\varkappa_\perp E+\Delta P_x$ and the independent variables of the problem will be ΔM_x, M_y, l_x, l_y, ΔP_x, P_y, and P_z. We also must use Eqs. (18) and (19). We then have

$$\tilde{F}_{MP}=\tfrac{1}{2}\chi^{-1}[\Delta M_x^2+(\chi H+\Delta M_x)^2 l_x^2+M_y^2]+\tfrac{1}{2}K(l_x^2+l_y^2)$$
$$-(\chi H+\Delta M_x)\left\{\gamma_1(\varkappa_\perp E l_y+P_y l_x)-l(\varkappa_\perp E\right.$$
$$\left.+\Delta P_x)\left[\gamma_2 l_x^2-\gamma_3\left(1-\frac{l_x^2+l_y^2}{2}\right)\right]\right\}-\gamma_0\chi H P_z l_x$$
$$+\tfrac{1}{2}\varkappa_\perp^{-1}(\Delta P_x^2+P_y^2)+\tfrac{1}{2}\varkappa_z^{-1}P_z^2. \quad (28)$$

Here we have again retained only terms of no higher than the second power in the γ_i (and also in l_x and l_y).

The procedure described for the previous case gives

$$\tilde{F}_{MP}=\tfrac{1}{2}(K_x l_x^2+K_y l_y^2), \quad (29)$$

where

$$K_x=K+\chi H^2[1+\chi\varkappa_\perp(\gamma_3(2\gamma_2+3\gamma_3)-\gamma_1^2)-\chi\varkappa_z\gamma_0^2]$$
$$+l(2\gamma_2+3\gamma_3)\chi\varkappa_\perp HE+\chi(\varkappa_\perp E)^2[2\gamma_3(\gamma_2$$
$$+\gamma_3)-\gamma_1^2],$$
$$K_y=K+\gamma_3 l\chi\varkappa_\perp HE+\chi\varkappa_\perp(\gamma_3^2-\gamma_1^2)(\chi H^2+\varkappa_\perp E^2).$$

Or, retaining only terms linear in the γ_i:

$$K_x=K+\chi H[H+(2\gamma_2+3\gamma_3)l\varkappa_\perp E], \quad (30)$$
$$K_y=K+\gamma_3 l\chi\varkappa_\perp HE. \quad (31)$$

After minimizing $\tilde{F}_{MP}+F_{le}$ with respect to l_x and l_y we find

$$l_x=-lK_x^{-1}(2B_{44}e_{xz}+2B_{41}e_{xy}), \quad (32)$$
$$l_y=-lK_y^{-1}[2B_{44}e_{yz}+B_{41}(e_{xx}-e_{yy})]. \quad (33)$$

Also

$$\Delta F_e=-\tfrac{1}{2}K_x^{-1}(2B_{44}e_{xz}+2B_{41}e_{xy})^2-\tfrac{1}{2}K_y^{-1}[2B_{44}e_{yz}$$
$$+B_{41}(e_{xx}-e_{yy})]^2.$$

This gives a renormalization of the following elastic moduli:

$$\tilde{C}_{66}=C_{66}-B_{41}^2/K_x,\quad \tilde{C}_{55}=C_{44}-B_{44}^2/K_x,$$
$$\tilde{C}_{44}=C_{44}-B_{44}^2/K_y,\quad \tilde{C}_{11}=C_{11}-B_{41}^2/K_y,$$
$$\tilde{C}_{12}=C_{12}+B_{41}^2/K_y. \quad (34)$$

Here, first of all, the isotropy of the elastic properties in the base plane disappears, since

$$\tilde{C}_{11}-\tilde{C}_{12}=2C_{66}-2B_{41}^2/K_y\neq 2\tilde{C}_{66}.$$

Secondly, there linear birefringence appears for waves with $\mathbf{k}\| \mathbf{z}$. Indeed, we have for the relative difference in the velocities of waves polarized along the x and the y axes

$$\frac{v_x-v_y}{v}=\frac{\tilde{C}_{55}-\tilde{C}_{44}}{2C_{44}}=\left(\frac{B_{44}}{K}\right)^2\frac{\chi H[H+2(\gamma_2+\gamma_3)l\varkappa_\perp E]}{2C_{44}}. \quad (35)$$

The magnitude of the effect depends both on H and on E. For one of the domains (e.g., the one with $l=+1$) it increases with increasing E, while for the other one ($l=-1$) it decreases, in contrast.

As to the connection with the antiferromagnetic resonance frequencies we have here

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

2. THE l⊥ z STATE

Let $\mathbf{H}\| \mathbf{x}$; for sufficiently large H overcoming the anisotropy in the base plane we then have $\mathbf{L}\| \mathbf{y}$. We consider the two cases for the direction of $\mathbf{E}$ which are the simplest as far as symmetry is concerned:

1) $\mathbf{E}\| \mathbf{H}\| \mathbf{x}(\mathbf{L}\| \mathbf{y})$

In this case $M_x=\chi H+\Delta M_x$ and $P_x=\varkappa_\perp E+\Delta P_x$ and for the independent variables we can take ΔM_x, M_z, l_x, l_z, ΔP_x, P_y, and P_z. We then have

$$M_y=-(M_x l_x+M_z l_z)/l_y\approx-(\chi H+\Delta M_x)l_x/l, \quad (36)$$
$$l_y\approx l-(l_x^2+l_z^2)/2l. \quad (37)$$

The expression for the thermodynamic potential $\tilde{F}_{MP}$ reduces in terms of those variables to the form:

$$\tilde{F}_{MP}=\tfrac{1}{2}\chi^{-1}[\Delta M_x^2+(\chi H+\Delta M_x)^2 l_x^2]+\tfrac{1}{2}Kl_z^2-(\chi H$$
$$+\Delta M_x)\left\{\left[\gamma_1 l\left(1-\frac{3l_x^2+l_y^2}{2}\right)+\gamma_3 l_z\right](\varkappa_\perp E\right.$$
$$\left.+\Delta P_x)+2\gamma_1 P_y l_x\right\}-\gamma_2 l M_z P_y+\tfrac{1}{2}\varkappa_\perp^{-1}(\Delta P_x^2$$
$$+P_y^2)+\tfrac{1}{2}\varkappa_z^{-1}P_z^2. \quad (38)$$

Minimizing it with respect to $\mathbf{M}$ and $\mathbf{P}$ gives

$$\tilde{F}_{MP}=\tfrac{1}{2}(K_x l_x^2+K_z l_z^2), \quad (39)$$

where

$$K_x=\chi H[H+5\gamma_1 l\varkappa_\perp E+\chi(4\gamma_1^2-\gamma_2^2)(\varkappa_\perp E)^2$$
$$\approx\chi H(H+5\gamma_1 l\varkappa_\perp E), \quad (40)$$
$$K_z=K+\gamma_1 l\chi\varkappa_\perp HE+(\gamma_1^2-\gamma_3^2)\chi\varkappa_\perp(\chi H^2+\varkappa_\perp E^2)$$
$$\approx K+\gamma_1 l\varkappa_\perp HE\chi. \quad (41)$$

The minimization of $\tilde{F}_{MP}$ of (39) with respect to l_x and l_z with the addition of the elastic energy

$$F_{le}=2(B_{44}l_z e_{yz}+B_{66}l_x e_{xy})l+[4B_{14}l_x e_{xz}+B_{41}l_z(e_{xx}$$
$$-e_{yy})]l$$

leads to the expressions

$$l_x=-K_x^{-1}(2B_{66}e_{xy}+4B_{14}e_{xz})l, \quad (42)$$

$$l_z=-K_z^{-1}[2B_{44}e_{yz}+B_{41}(e_{xx}-e_{yy})]l. \quad (43)$$

As a result we find the following renormalization of elastic moduli:

$$\begin{aligned}&\tilde{C}_{11}=C_{11}-B_{41}^2/K_z, \quad \tilde{C}_{12}=C_{12}+B_{41}^2/K_z,\\ &\tilde{C}_{55}=C_{44}-4B_{14}^2/K_x, \quad \tilde{C}_{44}=C_{44}-B_{44}^2/K_z,\\ &\tilde{C}_{66}=C_{66}-B_{66}^2/K_x,\\ &\tilde{C}_{14}=C_{14}-B_{41}B_{44}/K_z, \quad \tilde{C}_{65}=C_{14}-4B_{66}B_{14}/K_x.\end{aligned} \quad (44)$$

The quantities K_x from (40) and K_z from (41) determine, respectively, the low-frequency (ω_1) and the high-frequency (ω_2) antiferromagnetic resonance frequencies:

$$\omega_1^2=\gamma^2\chi^{-1}K_x, \quad \omega_2^2=\gamma^2\chi^{-1}K_z. \quad (45)$$

The magnetoelectric interaction thus makes according to (40), (41), and (45) additional contributions proportional to HE to the magnon gaps (in the approximation which is linear in the γ_i), while we must have $\gamma_1 l>0$ in the lowest ground state which corresponds to the highest excitation energy.

In this case the acoustic birefringence can become very strong since, by analogy with (35),

$$\frac{v_x-v_y}{v}=\frac{\tilde{C}_{55}-\tilde{C}_{44}}{2C_{44}}=\frac{B_{44}^2}{2K_zC_{44}}-\frac{2B_{14}^2}{K_xC_{44}}\approx-\frac{2B_{14}^2}{K_xC_{44}}. \quad (46)$$

The last, approximated part of the equation is written down under conditions where [see (40) and (41)]

$$K_x\ll K_z.$$

This can undoubtedly occur for sufficiently weak H and E fields. The situation is here completely analogous to the one for hematite (α-Fe_2O_3 and $FeBO_3$; see Ref. 11) only with the quantity

$$H_{ME}=5\gamma_1\varkappa_\perp E\equiv 5\chi^{-1}\alpha_{11}E \quad (47)$$

playing the role of the Dzyaloshinskiĭ field in this case.

2) $\mathbf{E}\| z(\mathbf{l}\| \mathbf{y},\mathbf{H}\| \mathbf{x})$

A calculation similar to the preceding case leads again to an expression of the form (39) where now

$$K_x=\chi H^2(1+\gamma_1^2\chi\varkappa_\perp)\approx\chi H^2,$$

$$K_z=K-\chi(\gamma_0\varkappa_z E)^2+(\gamma_1^2-\gamma_3^2)\varkappa_\perp(\gamma H)^2\approx K.$$

In this case there are no terms linear in the γ_i.

From the previous form of (42)–(45) we shall have formulas for l_x, l_z, $\omega_{1,2}$, and the $\tilde{C}_{ij}$ using the indicated substitution for K_x and K_z.

CONCLUSION

The main effects in acoustics caused by the magnetoelectric interaction are thus the following:

1) The magnetoelastic contribution to some of the elastic moduli becomes dependent on the electric field. This influence of $\mathbf{E}$ on the $\tilde{C}_{ij}$ can become particularly noticeable near orientational phase transitions where one of the acoustic modes softens.

2) The linear acoustic birefringence connected with magnetoelasticity also turns out to depend on $\mathbf{E}$.

3) Since these effects depend on the direction of $\mathbf{l}$ in the domains, there appears a possibility for an acoustic study of antiferromagnetic domain structure in magnetoelectric antiferromagnetics.

It follows fom Eqs. (40), (44), and (46) that these effects must manifest themselves most strongly in easy-plane antiferromagnetics in states for which $\mathbf{l}\perp z$ and $\mathbf{H}\| \mathbf{E}\| x$. In that case the effects (relative differences in the velocities or their change under the simultaneous action of the $\mathbf{E}$ and $\mathbf{H}$ fields) can apparently reach several times 10%. However, the author is unfortunataely not aware of any actual magnetoelectric antiferromagnetic of this kind which might be recommended for an experimental study.

As far as Cr_2O_3 is concerned, for which $\mathbf{l}\| z$, the situation turns out to be unfavorable. In order that the magnetoelectric contribution in (22) or (30) be comparable in order of magnitude, at least in the magnetic-field term, it is necessary to have a field $E\approx\alpha^{-1}\chi H\cdot 300$, if H is expressed in Oersted and E in V/cm. Taking[8] $\chi=10^{-3}$ and $\alpha=6\cdot 10^{-4}$ for Cr_2O_3 we find $E\approx 500\,H$. For more rigorous estimates we must perform the calculations using a model in which $\chi_\| \neq 0$.

In a subsequent paper we shall consider tetragonal magnetoelectric antiferromagnetics with $\bar{1}\ 4_z^+2_d^-$ and $\bar{1}\ 4_z^-2_d^-$ structures. This is necessary because, firstly, a whole series of magnetoelectrics with such structures is known, both easy-axis and easy-plane ones. Secondly, the magnetoelectric antiferromagnetic with the largest magnetoelectric susceptibility $\alpha\approx 1\cdot 10^{-2}$ ($TbPO_4$) is just one of them. Moreover, it is interesting to elucidate the different properties for antiferromagnetics with different parities of the 4_z symmetry axis. (The 3_z axis can only be even.)

Finally, a comparison of the results for those tetragonal magnetoelectrics with the results of the present paper for the $\bar{1}\ 3_z^+2_x^-$ structure makes it possible to give a rather complete answer for the following problems, of interest to us, of the acoustics of antiferromagnetic magnetoelectrics: Which qualitative regularities of the effect of the magnetoelectric interaction on the acoustic properties distinguish antiferromagnetics with different magnetic structures and states? Which effects is it especially desirable to study experimentally, and in what actual materials? And finally, what useful information can magneto-electro-acoustics give about magnetoelectric antiferromagnetics?

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