Elastic anharmonicity in magnetoelectric tetragonal antiferromagnets

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For tetragonal antiferromagnets that have an asymmetry center and exhibit a magnetoelectric effect we examine the effective elastic anharmonicity of magnetoelastic origin, allowing for this effect. We show that in some cases the magnetoelastic and magnetoelectric interactions lead to new effective nonlinear elasticity moduli, which are finite only when magnetic and electric fields are applied to the sample simultaneously. We find the conditions under which the elastic anharmonicity associated with antiferromagnetism becomes gigantic and strongly dependent on the applied magnetic and electric fields. Usually this occurs near magnetic orientational phase transitions.

1. INTRODUCTION

The features of the linear acoustic properties of tetragonal antiferromagnets that allow for the existence of a magnetoelectric effect have been studied in Ref. 1. The velocity of elastic waves and the acoustic double refraction were found to depend on the magnetic and electric fields. The present paper develops these investigations and studies the nonlinear acoustic properties of such antiferromagnets. As in Ref. 1, we consider only low frequencies,

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

where ω_{AFMR} is the lowest antiferromagnetic resonance (AFMR) frequency. Condition (1) makes it possible, without solving the equations of magnetoelastic dynamics, to find the renormalization related to magnetoelastic and magnetoelectric interactions of not only linear elasticity moduli but also nonlinear elasticity moduli. To this end one must employ the fact that at such frequencies the antiferromagnetism vector **L**, the magnetization vector **M**, and the polarizability vector **P** follow the elastic strains e_{ij} in a quasiequilibrium manner.

The calculations are carried out using the model of two sublattices with constant absolute values of the magnetizations,

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

in which the longitudinal (in relation to the antiferromagnetism vector) magnetic susceptibility $\chi_\parallel$ is zero. The model is applicable only for fairly low temperatures.

2. THE THERMODYNAMIC POTENTIAL

Let us examine tetragonal antiferromagnets that manifest themselves in the following centrally symmetric exchange magnetic structures:[2]

$$\bar{1}(-)4_z(+)2_d(-) \equiv \bar{1}(-)4_z(+)2_x(-),$$

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

These structures are characteristic of a large number of trirutiles (Fe_2TeO_6, etc.) and rare-earth phosphates ($HoPO_4$, etc.) and vanadates ($GdVO_4$, etc.); see the literature cited in Ref. 1.[1)]

The total thermodynamic potential density,

$$F = F_m + F_p + F_e + F_{le} + F_{mp}, \tag{3}$$

consists of the magnetic contribution F_m, the electropolarization contribution F_p, the elastic contribution F_e, the antiferromagnetic contribution F_{le}, and the magnetoelectric contribution F_{mp}:

$$F_m = \frac{1}{2}\chi^{-1}M^2 - \mathbf{MH} + \frac{1}{2}K(l_x^2 + l_y^2)\left(\text{or } \frac{1}{2}Kl_z^2\right) + \frac{1}{2}K_2 l_x^2 l_y^2,$$

$$F_p = \frac{1}{2}\kappa_\perp^{-1}(P_x^2 + P_y^2) + \frac{1}{2}\kappa_\parallel^{-1}P_3^2 - \mathbf{PE},$$

$$F_e = \frac{1}{2}C_{11}(e_{xx}^2 + e_{yy}^2) + C_{12}e_{xx}e_{yy} + C_{13}(e_{xx} + e_{yy})e_{zz} + \frac{1}{2}C_{33}e_{zz}^2 + 2C_{44}(e_{xz}^2 + e_{yz}^2) + 2C_{66}e_{xy}^2 + \frac{1}{6}C_{ijklmn}e_{ij}e_{kl}e_{mn},$$

$$F_{le} = B_{11}(l_x^2 e_{xx} + l_y^2 e_{yy}) + B_{12}(l_x^2 e_{yy} + l_y^2 e_{xx}) + B_{13}(e_{xx} + e_{yy})l_z^2 + B_{31}e_{zz}(l_x^2 + l_y^2) + B_{33}e_{zz}l_z^2 + 2B_{44}l_z(e_{xz}l_x + e_{yz}l_y) + 2B_{66}e_{xy}l_x l_y. \tag{4}$$

The nonlinear elasticity moduli C_{ijklmn} in (4) for tetragonal crystals are listed in Ref. 4. The magnetoelectric contributions as functions of the parity of the magnetic structure with respect to the 4_z axis have the form[1]

$$(4_z(+))F_{mp} = -\gamma_2(l_x P_x + l_y P_y)M_z - \gamma_3(M_x P_x + P_y M_y)l_z - \gamma_4(l_x M_x + l_y M_y)P_z$$

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$$-\gamma_5 l_z M_z P_z, \quad (5)$$

$$(4_z(-))F_{mp} = -\gamma_2(l_x P_y + l_y P_x)M_z - \gamma_3(M_x P_y + M_y P_x)l_z - \gamma_4(M_x l_y + M_y l_x)P_z, \quad (6)$$

with $\gamma_i (i= 2-5)$ the magnetoelectric constants, which determine the magnetoelectric susceptibility: $\alpha = \chi\kappa\gamma$ (without indices), where χ and κ are the magnetic and dielectric susceptibilities, respectively.

Our goal is to establish the influence of the magnetoelastic effect on the anharmonicity of the specified antiferromagnets as a function of the magnetic state (easy axis and easy plane) for different orientations of the magnetic and electric fields.

We describe the idea behind the calculation, using the first of these cases as an example.

3. THE EXCHANGE MAGNETIC STRUCTURE $\bar{1}(-)4_z(-)2_d(-)$

3.1. Easy axis: H ∥ E ∥ L ∥ z

Owing to condition (2) only seven independent dynamical variables remain, for which in the given case we select the following: M_x, M_y, P_x, P_y, P_z, l_x, and l_y.

We start by minimizing the quantity

$$\tilde{F}_{mp} = F_m + F_p + F_{mp} \quad (7)$$

in **P**, M_x, and M_y, which enables expressing the vectors **P** and **M** in terms of **l**. Substituting the obtained expressions into (6), we arrive at the following result for $\tilde{F}_{mp}$:

$$\tilde{F}_{mp} = -\kappa_\parallel \gamma_4 E(l_y M_x + l_x M_y) + \frac{H}{l_z}(M_x l_x + M_y l_y) + \frac{1}{2}M_x^2 R_1 + \frac{1}{2}M_y^2 R_2 + M_x M_y R_3 l_z^2 + \frac{1}{2}K(l_x^2 + l_y^2) + \frac{1}{2}K_2 l_x^2 l_y^2 + \frac{H}{2l_z}(l_x^2 + l_y^2)(M_x l_x + M_y l_y), \quad (8)$$

where

$$M_x = \left[\left(\kappa_\parallel \gamma_4 l_y E - H\frac{l_x}{l_z}\right)R_2 - \left(\kappa_\parallel \gamma_4 l_x E - H\frac{l_y}{l_x}\right)R_3\right] \times \frac{1}{R_1 R_2 - R_3^2},$$

$$R_1 = \frac{1}{\chi}\left(1 + \frac{l_x^2}{l_y^2}\right), \quad R_2 = R_1(\text{ as } l_x \to l_y), \quad R_3 = \frac{l_x l_y}{\chi l_z^2}, \quad (9)$$

$$M_y = M_x(\text{ as } l_x \to l_y), \quad l_z^2 \approx 1 - l_x^2 - l_y^2.$$

In both Eqs. (8) and (9) we discard the terms in the R_i ($i=1$, 2, 3) that are proportional to $\gamma_i \gamma_j \kappa_{\parallel,\perp}$, in view of their smallness.

Below, when discussing nonlinear elasticity moduli through third order, we must write $\tilde{F}_{mp}$ in an approximation quadratic in γ_i to within fourth-order terms in l_x and l_y. Note that in this case the components l_x and l_y do not belong to different sets of normal coordinates (these are the variables $l_{x'} = (l_x + l_z)/\sqrt{2}$ and $l_{y'} - (-l_x + l_y)/\sqrt{2}$). Hence it is convenient to transfer to a system of coordinates (x', y', z) rotated about the z axis by 45°. In the "primed" system of coordinates we arrive at the following expression for $\tilde{F}_{mp} + F_{le}$:

$$\tilde{F}_{mp} + F_{le} = \frac{1}{2}K_x l_{x'}^2 + \frac{1}{2}K_y l_{y'}^2 + 2B_{44} l_z(e_{x'z} l_{x'} + e_{y'z} l_{y'}) + \frac{K_2}{4}(l_{x'}^2 - l_{y'}^2)^2 - \frac{1}{2}\alpha E H l_z^0 (l_{x'}^4 - l_{y'}^4) + \frac{3}{2}\chi H^2 (l_{x'}^2 + l_{y'}^2)^2 + \frac{1}{2}B_{11}^{(1)}(l_{x'}^2 e_{x'x'} + l_{y'}^2 e_{y'y'}) + \frac{1}{2}B_{12}^{(1)}(l_{y'}^2 e_{x'x'} + l_{x'}^2 e_{y'y'}) + B_{31}^{(1)} e_{zz}(l_{x'}^2 + l_{y'}^2) + 2B_{66}^{(1)} l_{x'} l_{y'} e_{x'y'}, \quad (10)$$

where we have introduced the notation

$$\begin{aligned} K_x &= K - \chi(H - \kappa_\parallel \gamma_4 E l_z^0)^2, \\ K_y &= K - \chi(H + \kappa_\parallel \gamma_4 E l_z^0)^2, \\ B_{11}^{(1)} &= B_{11} + B_{12} + B_{66} - 2B_{13}, \\ B_{12}^{(1)} &= B_{11} + B_{12} - 2B_{13} - B_{66}, \\ B_{31}^{(1)} &= B_{31} - B_{33}, \quad B_{66}^{(1)} = B_{11} - B_{12}, \\ \alpha &= \chi \gamma_4 \kappa_\parallel, \end{aligned} \quad (11)$$

and $l_z^0 = +1$ or -1 for domains with **l**↑↑**z** and **l**↑↓**z**. In Eq. (10) in terms containing the fourth powers of the components $l_{x'}$ and $l_{y'}$ we ignored the terms of the types $\chi\gamma_i\gamma_j H^2 \kappa_{\parallel,\perp}$ and $\chi\kappa_\parallel^2 \gamma_\parallel^2 E^2$ because of their smallness. Minimizing then (10) in $l_{x'}$ and $l_{y'}$, we express these variables in terms of the strains e_{ij} that generate them in a quadratic approximation:

$$\begin{aligned} l_{x'} &= -\frac{2B_{44}}{K_x} l_z^0 e_{x'z}\left(1 - e_{x'x'}\frac{B_{11}^{(1)}}{K_x} - e_{y'y'}\frac{B_{12}^{(1)}}{K_x}\right) + \frac{4B_{31}^{(1)} B_{44}}{K_x^2} e_{x'z} e_{zz} l_z^0 + \frac{4B_{66}^{(1)} B_{44}}{K_x K_y} l_z^0 e_{y'z} e_{x'y'}, \\ l_{y'} &= -\frac{2B_{44}}{K_y} l_z^0 e_{y'z}\left(1 - e_{x'x'}\frac{B_{12}^{(1)}}{K_y} - e_{y'y'}\frac{B_{11}^{(1)}}{K_y}\right) + \frac{4B_{31}^{(1)} B_{44}}{K_y^2} l_z^0 e_{y'z} e_{zz} + \frac{4B_{66}^{(1)} B_{44}}{K_x K_y} l_z^0 e_{x'z} e_{x'y'}. \end{aligned} \quad (12)$$

Equations (11) were obtained in the approximation $B_{\alpha\beta}^{(1)} e_{ij} \ll K_x\ K_y$.

Substituting Eqs. (12) into (10) and returning to the initial system of coordinates, we find that here the nonlinear elasticity moduli C_{144}, C_{155}, C_{244}, C_{255}, C_{344}, C_{355}, and C_{456} are renormalized and that the following new nonzero effective elasticity moduli emerge: $\tilde{C}_{154}$, $\tilde{C}_{254}$, $\tilde{C}_{354}$, $\tilde{C}_{644}$, and C_{655}. Here and in what follows we introduce the notation $C_{\alpha\beta\gamma}$ for third-order elasticity moduli, where α, β, and γ label in the usual way[4] from 1 to 6 the pairs of indices i,j, each of which varies from 1 to 3. Below we also assume that the renormalized nonlinear moduli can be written in the form

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

where the terms $\Delta C_{\alpha\beta\gamma}(\mathbf{E},\mathbf{H})$ determine the renormalization caused by magnetoelastic and magnetoelectric interactions. In the case at hand for $\Delta C_{\alpha\beta\gamma}$ we obtain

$$\Delta C_{144}(\mathbf{E},\mathbf{H})=-\frac{12(B_{11}-B_{12})B_{44}^2}{\chi^2H_1^2H_2^2}+\frac{6(B_{11}+B_{12}-2B_{13})B_{44}^2}{\chi^4H_1^4H_2^4}(K_x^2+K_y^2)$$

$$=\Delta C_{255}(\mathbf{E},\mathbf{H}),$$

$$\Delta C_{244}(\mathbf{E},\mathbf{H})=\Delta C_{155}(\mathbf{E},\mathbf{H})=\Delta C_{144}(\mathbf{E},\mathbf{H})+\frac{24(B_{11}-B_{12})B_{44}^2}{\chi^2H_1^2H_2^2},$$

$$\Delta C_{344}(\mathbf{E},\mathbf{H})=C_{355}(\mathbf{E},\mathbf{H})=\frac{12(B_{11}-B_{33})B_{44}^2}{\chi^4H_1^4H_2^4}\times(K_x^2+K_y^2),$$

$$\Delta C_{456}(\mathbf{E},\mathbf{H})=\frac{24B_{66}B_{44}^2}{\chi^4H_1^4H_2^4}(K_x^2+K_y^2), \qquad (13)$$

for $\tilde{C}_{\alpha\beta\gamma}$ we have

$$\tilde{C}_{154}=\Delta C_{154}(\mathbf{E},\mathbf{H})=\Delta C_{254}(\mathbf{E},\mathbf{H})=\tilde{C}_{254}$$

$$=-\frac{96(B_{11}+B_{12}-2B_{13})B_{44}^2}{\chi^3H_1^4H_2^4}\gamma_4\kappa_\parallel EHl_z^0\{K-\chi[H^2+(\gamma_4\kappa_\parallel W)^2]\}, \qquad (14)$$

and the moduli $\tilde{C}_{644}=\tilde{C}_{655}$ and $\tilde{C}_{354}$ can be obtained from $\tilde{C}_{154}$ (Eq. (14)) by replacing $B_{11}+B_{12}-2B_{13}$ with B_{66} and $2(B_{31}-B_{33})$. Here we have introduced the notation $H_{1,2}=\omega_{1,2}/g$, where g is the gyromagnetic ratio, and ω_1 and ω_2 are the AFMR frequencies determined by the following expressions:[1]

$$\omega_{1,2}^2=g^2[(\sqrt{K\chi^{-1}}\mp H)^2-(\alpha\chi^{-1}E)^2].$$

As Eqs. (14) show, the new effective nonlinear elasticity moduli $\tilde{C}_{154}$, etc., are nonzero only if the magnetic and electric fields are applied to the sample simultaneously, i.e., these moduli result from the magnetoelectric interaction. Formally, the new moduli arise because the effective anisotropy constants K_x and K_y (Eq. (11)) depend differently on the electric and magnetic fields. As shown in Ref. 1, this difference within the given geometry occurs only in antiferromagnets with an exchange magnetic structure that is odd with respect to the 4_z axis and is related to a definite form of the magnetoelectric terms (6) in the thermodynamic potential. The condition for the stability of the state with $\mathbf{l}\parallel\mathbf{z}$ can be formulated in terms of the following inequalities:

$$\tilde{C}_{44}=C_{44}-\frac{B_{44}^2}{K_y}\geqslant 0, \quad \tilde{C}_{55}=C_{44}-\frac{B_{44}^2}{K_x}\geqslant 0. \qquad (15)$$

This together with (11) implies that the threshold of stability in the fields $\mathbf{E}$ and $\mathbf{H}$ is determined by the equation

$$(H+|\alpha|\chi^{-1}E)^2=\chi^{-1}\left(K-\frac{B_{44}^2}{C_{44}}\right). \qquad (16)$$

Near this threshold the antiferromagnetic modulus $\tilde{C}_{154}$ proves to be of the following order of magnitude:

$$\tilde{C}_{154}(\mathbf{E},\mathbf{H})=-\frac{96|\alpha|EHl_z^0(B_{11}+B_{12}-2B_{13})C_{44}^3}{B_{44}^2(B_{44}^2+4|\alpha|EHl_z^0C_{44})^2}\times(B_{44}^2+2|\alpha|EHl_z^0C_{44}). \qquad (17)$$

If we assume that $B_{\alpha\beta}\sim 10^6$ erg cm^{-3}, $C_{44}\sim 3\times 10^{11}$ erg cm^{-3}, $H\sim 10^4$Oe, $E\sim 10$ esu, and $\alpha\sim 10^{-4}$, then

$$|\tilde{C}_{154}|\sim 10^{18}\text{erg cm}^{-3}.$$

For comparison we note that the ordinary anharmonicity of crystals is of order 10^{11}–10^{13}erg cm^{-3}.

3.2. Easy plane: L ∥ y, H ∥ x, and E ∥ z

Since the procedure for calculating the renormalization of nonlinear elasticity moduli is described above rather thoroughly, we list only the results in what follows.

For the antiferromagnet of the easy-plane type discussed here it is convenient to take l_x, l_z, M_x, M_z, P_x, P_y, P_z, and e_{ij} as the independent variables.

The expression for $\tilde{F}_{mp}$ in the approximation that is linear in γ_i and up to fourth order in l_i $(i=x,z)$ can be written as follows:

$$\tilde{F}_{mp}=\frac{1}{2}K_xl_x^2+\frac{1}{2}K_zl_z^2+\frac{1}{2}Q_1l_x^4+\frac{1}{2}Q_2l_z^4+\frac{1}{2}Q_3l_x^2l_z^2, \qquad (18)$$

where

$$K_z=K+\chi\gamma_4\kappa_\parallel l_y^0EH,$$

$$K_x=K_2+5\chi\gamma_4\kappa_\parallel l_y^0EH+\chi H^2,$$

$$Q_1=-K_2-\chi H^2+4\chi\gamma_4\kappa_\parallel EHl_y^0-8\chi\gamma_4\kappa_\parallel EHl_y^0, \qquad (19)$$

$$Q_2=-\chi H^2+\chi\gamma_4\kappa_\parallel EHl_y^0,$$

$$Q_3=-K_2+\chi H^2+6\chi\gamma_4EHl_y^0$$

($l_y^0=\pm 1$ for 180-degree domains). Now we assume $l_y^0=1$. Then

$$l_x=-\frac{2B_{66}}{K_x}e_{xy}\left[1-\frac{2(B_{11}-B_{12})}{K_x}e_{xx}-\frac{2(B_{12}-B_{11})}{K_x}e_{yy}\right]+\frac{4B_{44}^2}{K_xK_z}e_{xx}e_{yz},$$

$$l_z=-\frac{2B_{44}}{K_z}e_{yz}\left[1-\frac{2(B_{13}-B_{12})}{K_z}e_{xx}-\frac{2(B_{13}-B_{11})}{K_z}e_{yy}-\frac{2(B_{33}-B_{31})B_{66}}{K_z}e_{zz}\right]+\frac{4B_{44}^2}{K_xK_z}e_{xz}e_{yz}. \qquad (20)$$

As a result the elastic moduli C_{166}, C_{266}, C_{144}, C_{244}, C_{344}, and C_{456} become renormalized, so that

$$\Delta C_{166}(\mathbf{E,H}) = -\Delta C_{266}(\mathbf{E,H}) = \frac{24(B_{11}-B_{12})}{\chi^2 H_1^4} B_{66}^2,$$

$$\Delta C_{144}(\mathbf{E,H}) = -\frac{24(B_{12}-B_{13})}{\chi^2 H_2^4} B_{44}^2,$$

$$\Delta C_{244}(\mathbf{E,H}) = -\frac{24(B_{11}-B_{13})}{\chi^2 H_2^4} B_{44}^2, \quad (21)$$

$$\Delta C_{344}(\mathbf{E,H}) = \frac{24(B_{33}-B_{31})B_{44}^2}{\chi^2 H_2^4},$$

$$\Delta C_{456}(\mathbf{E,H}) = \frac{48 B_{66} B_{44}^2}{\chi^2 H_1^4 H_2^4},$$

where

$$H_1^2 = \frac{\omega_1^2}{g^2} = \frac{K_x}{\chi}, \quad H_2^2 = \frac{\omega_2^2}{g^2} = \frac{K_z}{\chi}, \quad (22)$$

where ω_1 and ω_2 are the lower and upper AFMR frequencies.[1] The most strongly renormalized moduli are C_{166} and C_{266}, which are related to ω_1. The stability criterion for the state considered here is the requirement that the squares of the acoustic modes with $\mathbf{k}\|\mathbf{y}$ be nonnegative, which determines the lowest possible values of the effective anisotropy constants:

$$K_x \geq \frac{B_{66}^2}{C_{66}}, \quad K_z \geq \frac{B_{44}^2}{C_{44}}.$$

Hence in the vicinity of an orientational transition in the basal plane (where $K_x \to B_{66}^2/C_{66}$), the value of $\Delta C_{166}(\mathbf{E,H})$ may become huge:

$$\Delta C_{166}(\mathbf{E,H}) \approx \frac{24(B_{11}-B_{12})}{B_{66}^2} C_{66}^2 \sim 10^{18}\ \text{erg/cm}^{-3}.$$

Note that the above transition is achieved most easily for $K_2<0$ (for the base anisotropy stated in Eq. (4)), when the direction $\mathbf{H}\|\mathbf{x}$ is the difficult one in the basal plane and we approach the transition point from the range of values of H satisfying the condition

$$\chi H^2 + 5\alpha E H > \frac{B_{66}^2}{C_{66}} + |K_2|. \quad (23)$$

In this geometry not only is the renormalization of the corresponding nonlinear moduli the greatest but the moduli also exhibit a strong dependence on the fields **E** and **H**.

As is well known,[4] in tetragonal crystals the nonlinear moduli C_{344} and C_{355} must be equal if the magnetoelastic and magnetoelectric interactions are to be ignored. The inequalities (21) imply that only C_{344} is renormalized as a result of these interactions, while the nonlinear modulus C_{355} remains unchanged. This means that in easy-plane antiferromagnets the magnetoelectric and magnetoelastic interactions disrupt the symmetry in the relationships between nonlinear moduli rather than giving rise to new nonlinear moduli.

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

4.1. Easy axis: E ∥ H ∥ L ∥ z

The independent variables are the same as in Sec. 3.1. To second order in the γ_i, the thermodynamic potential $\tilde{F}_{mp}$ can be written as

$$\tilde{F}_{mp} = \frac{1}{2}\tilde{K}(l_x^2+l_y^2) + \frac{1}{2}\chi(H-\gamma_0\kappa_\| E l_z^0)^2(l_x^2+l_y^2)^2 + \frac{1}{2}K_2 l_x^2 l_y^2, \quad (24)$$

where

$$\tilde{K} = K - \chi(H-\kappa_\|\gamma_0 l_z^0 E)^2, \quad \gamma_0 = \gamma_4 - \gamma_5. \quad (25)$$

In Eq. (24) we discarded the terms of type $\kappa_\|\gamma_i\gamma_j(H-\kappa_\|\gamma_0 l_z^0 E)^2[*](l_x^2+l_y^2)^2$ in view of their smallness. For l_x and l_y we have

$$l_x = -\frac{2B_{44}}{\tilde{K}} l_z^0 e_{xz}\left[1 - \frac{2(B_{11}-B_{13})}{\tilde{K}} e_{xx} - \frac{2(B_{12}-B_{13})}{\tilde{K}} e_{yy} - \frac{2(B_{31}-B_{33})}{\tilde{K}} e_{zz}\right] + \frac{4B_{44}B_{66}}{\tilde{K}^2} e_{yz} e_{xy} l_z^0$$

$$l_y = -\frac{2B_{44}}{\tilde{K}} l_z^0 e_{yz}\left[1 - \frac{2(B_{11}-B_{13})}{\tilde{K}} e_{yy} - \frac{2(B_{12}-B_{13})}{\tilde{K}} e_{xx} - \frac{2(B_{31}-B_{33})}{\tilde{K}} e_{zz}\right] + \frac{4B_{44}B_{66}}{\tilde{K}^2} e_{xz} e_{xy} l_z^0. \quad (26)$$

And again, as in Eqs. (24)–(26), $l_z^0 = \pm 1$ depending on the direction of **l** in the domain. In what follows we take a domain with $l_z^0 = 1$.

Bearing in mind Eqs. (25), we find that the nonlinear moduli C_{144}, C_{244}, C_{155}, C_{255}, C_{344}, C_{355}, and C_{456} are renormalized, with

$$\Delta C_{144}(\mathbf{E,H}) = \Delta C_{255}(\mathbf{E,H}) = \frac{24(B_{12}-B_{13})B_{44}^2}{\chi^2 H_1^2 H_2^2},$$

$$\Delta C_{244}(\mathbf{E,H}) = \Delta C_{155}(\mathbf{E,H}) = \frac{24(B_{11}-B_{13})B_{44}^2}{\chi^2 H_1^2 H_2^2}, \quad (27)$$

$$\Delta C_{344}(\mathbf{E,H}) = \Delta C_{355}(\mathbf{E,H}) = \frac{24(B_{31}-B_{33})B_{44}^2}{\chi^2 H_1^2 H_2^2},$$

$$\Delta C_{456}(\mathbf{E,H}) = \frac{48 B_{66} B_{44}^2}{\chi^2 H_1^2 H_2^2}.$$

The AFMR frequencies ω_1 and ω_2 satisfy the following relationships:[1]

$$H_1H_2=\frac{\omega_1\omega_2}{g^2}=\frac{\tilde{K}}{\chi}.$$

Note that no new effective dynamical moduli emerge here, in contrast to antiferromagnets with an exchange magnetic structure odd with respect to the fourth-order symmetry axis.

Equations (27) imply that effective dynamical moduli increase significantly near spin–reorientation transitions. For instance, since the condition for magnetoelastic stability of the state in question is the inequality

$$\tilde{K}\geqslant\frac{B_{44}^2}{C_{44}}, \tag{28}$$

the asymptotic value of the moduli C_{155} and C_{244} may reach values of order

$$\Delta C_{155}(\mathbf{E},\mathbf{H})=\Delta C_{244}(\mathbf{E},\mathbf{H})\approx 24\frac{(B_{11}-B_{13})}{B_{44}^2}C_{44}^2$$
$$\sim 10^{18}\text{erg/cm}^{-3} \tag{29}$$

if for the purpose of making an estimate we again assume $B_{\alpha\beta}\sim 10^6\text{erg/cm}^{-3}$ and $C_{44}\sim 3\times 10^{11}\text{erg/cm}^{-3}$. But observing renormalizations of this order requires applying strong fields to the sample, so that in accordance with (28)

$$\sqrt{\left(K-\frac{B_{44}^2}{C_{44}}\right)\frac{1}{\chi}}\approx H-\gamma_0\kappa_{\parallel}E. \tag{30}$$

Assuming that $K\sim 10^5\text{erg/cm}^{-3}$ and $\chi\sim 10^{-3}$, we obtain $H\sim 10^4$ Oe.

4.2. Easy plane: L ∥ y, H ∥ z, and E ∥ y

As in Sec. 3.2, we take l_x, l_z, M_x, M_z, P_x, P_y, P_z, and e_{ij} as the dynamical variables of the problem. Here the potential $\tilde{F}_{mp}$ can be examined in an approximation that is linear in γ_i, since acoustic magnetoelectric effects appear even in this approximation. As a result we have

$$\tilde{F}_{mp}=\frac{1}{2}(K_zl_z^2+K_xl_x^2+K_{xx}l_x^4+K_{xz}l_x^2l_z^2+K_{zz}l_z^4), \tag{31}$$

where

$$\begin{aligned} &K_x=K_2+\chi\gamma_2\kappa_\perp EHl_y^0,\\ &K_z=K+\chi H^2+\chi\kappa_\perp(3\gamma_2+2\gamma_3)EHl_y^2,\\ &K_{xx}=-K_2,\quad K_{xz}=\chi H^2-2\gamma_2\kappa_\perp EHl_y^2-K_2,\\ &K_{zz}=-\chi H^2-2\chi\kappa_\perp(\gamma_2+3\gamma_1)EHl_y^0. \end{aligned} \tag{32}$$

In antiferromagnets of the type considered here the renormalized elastic moduli are the same as in Sec. 3.2. For the $\Delta C_{\alpha\beta\gamma}(\mathbf{E},\mathbf{H})$ we have expressions similar to (21) with AFMR frequencies (or $H_{1,2}$) corresponding to the effective anisotropy constants K_x and K of Eqs. (32). All that was said in Sec. 3.2 concerning the conditions and values of the renormalizations of elastic moduli remains valid. Note also that here (and in Sec. 3.2, for that matter) the orientational transition in the basal plane can occur along the field **E** for a given **H**.

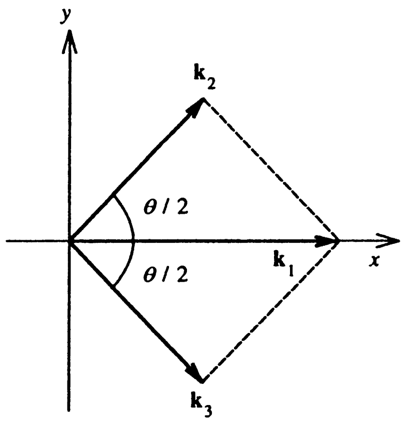

FIG. 1.

5. EXAMPLES OF SCATTERING PROCESSES

Here are some examples of nonlinear effects related to the anharmonic moduli $\tilde{C}_{\alpha\beta\gamma}$.

5.1. The process related to $\tilde{C}_{144}$, $\tilde{C}_{155}$, and $\tilde{C}_{154}$

According to Eqs. (13) and (14), this process is possible for an antiferromagnet with the exchange magnetic structure $\bar{1}(-)4_z(-)2_d(-)$ in a state with $\mathbf{L}\parallel\mathbf{z}$. What is important here is that $\tilde{C}_{144}$, $\tilde{C}_{155}$, $\tilde{C}_{154}$ depend on both **E** and **H**, with the modulus $\tilde{C}_{154}$ caused by the magnetoelectric interaction and finite only if $EH\neq 0$. For example, this can be the decay of a longitudinal phonon with a wave vector $\mathbf{k}_1\parallel\mathbf{x}$ into two transverse phonons with polarizations $\mathbf{u}_2\parallel\mathbf{u}_3\parallel\mathbf{z}$, frequencies ω_2 and ω_3, and wave vectors $\mathbf{k}_2$ and $\mathbf{k}_3$ lying in the xy plane at the same angles $\theta/2$ to the x axis (Fig. 1). With such directions of phonon propagation we find that

$$\begin{aligned} &k_{2x}=k_{3x}=k\cos\frac{\theta}{2},\\ &k_{2y}=-k_{3y}=k\sin\frac{\theta}{2},\\ &k=|\mathbf{k}_2|=|\mathbf{k}_3|, \end{aligned} \tag{33}$$

and the corresponding frequencies determined from the equations of the elastic dynamics for the incident (ω_1) and scattered (ω_2 and ω_3) waves have the form

$$\begin{aligned} &\omega_1=k_1\sqrt{\frac{C_{11}}{\rho_0}},\\ &\omega_{2,3}^2=k^2\frac{\tilde{C}_{44}+\tilde{C}_{55}-(\tilde{C}_{44}-\tilde{C}_{55})\sin\theta}{2\rho_0}, \end{aligned} \tag{34}$$

where ρ_0 is the density of the medium prior to strain, and $\tilde{C}_{44}$ and $\tilde{C}_{55}$ are determined by Eqs. (15) together with (11).

Equations (33) and (34) imply that the synchronism conditions

$$\omega_1=\omega_2+\omega_3,\quad \mathbf{k}_1=\mathbf{k}_2+\mathbf{k}_3, \tag{35}$$

which represent the laws of conservation of phonon energy and quasimomentum, can be met in such a geometry if

$$\omega_{2,3}=\frac{\omega_1}{2}\left(1\mp\frac{\tilde{C}_{44}-\tilde{C}_{55}}{C_{11}}\sqrt{\frac{2C_{11}-(\tilde{C}_{44}+\tilde{C}_{55})}{\tilde{C}_{44}+\tilde{C}_{55}}}\right),$$
$$\cos^2\frac{\theta}{2}=\frac{\tilde{C}_{44}+\tilde{C}_{55}}{2C_{11}},\quad k=\frac{k_1}{2}\sqrt{\frac{2C_{11}}{\tilde{C}_{44}+\tilde{C}_{55}}}. \qquad (36)$$

Here the difference in the frequencies ω_2 and ω_3, in accordance with Eqs. (15) and (11), is due entirely to the magnetoelectric interaction, which leads to $\tilde{C}_{44}\neq\tilde{C}_{55}$. Equations (36) were obtained in an approximation where $C_{11}\gg\tilde{C}_{44}-\tilde{C}_{55}$. The dependence of the anharmonic processes on **E** and **H**, a characteristic feature of magnetoelectric antiferromagnets, can be seen even in (36). This is especially true in the region of orientational phase transitions. Moreover, the dependence of the moduli $\tilde{C}_{\alpha\beta\gamma}$ on **E** and **H** and their huge size is highly important for the intensity of these processes.

Far from the region of the spin-flop transition, where

$$C_{44}\gg\frac{B_{44}^2}{K_y},\quad C_{44}\gg\frac{B_{44}^2}{K_x},$$

we have $\cos^2(\theta/2)\approx C_{44}/C_{11}$ and

$$\omega_{2,3}=\frac{\omega_1}{2}\left(1\pm\frac{4|\alpha|EHB_{44}^2}{\chi^2H_1^2H_2^2C_{11}}\sqrt{\frac{C_{11}-C_{44}}{C_{44}}}\right). \qquad (37)$$

Here we have allowed only for the largest terms dependent on **E** and **H**. Equation (37) shows that one of the frequencies, ω_2 or ω_3, increases linearly with the electric field strength (at a fixed value of H) while the other decreases.

In the asymptotic region of the spin-flop transition, where $\tilde{C}_{44}$ or $\tilde{C}_{55}$ tends to zero, the angle of divergence of the generated phonons can be found from the condition that

$$\cos^2\frac{\theta}{2}=\frac{2|\alpha|EHC_{44}^2}{C_{11}(B_{44}^2+4|\alpha|EHC_{44})}, \qquad (38)$$

and the frequencies ω_2 and ω_3 and the absolute value of the vectors $\mathbf{k}_2$ and $\mathbf{k}_3$ are determined by the following relationships:

$$\omega_{2,3}=\frac{\omega_1}{2}\left(1\pm\sqrt{\frac{8|\alpha|EHC_{44}^2[C_{11}B_{44}^2+2|\alpha|EHC_{44}(2C_{11}-C_{44})]}{C_{11}^2(B_{44}^2+4|\alpha|EHC_{44})^2}}\right),$$
$$k=\frac{1}{2}k_1\sqrt{\frac{C_{11}(B_{44}^2+4|\alpha|EHC_{44})}{2|\alpha|EHC_{44}^2}}, \qquad (39)$$

where the values of the external electric (**E**) and magnetic (**H**) fields in Eq. (38) must satisfy the condition (16). If an orientational phase transition is achieved at such values of E and H that $B_{44}^2\gg4|\alpha|EHC_{44}$, then

$$\omega_{2,3}=\frac{\omega_1}{2}\left(1\pm\sqrt{\frac{8|\alpha|EHC_{44}^2}{C_{11}B_{44}^2}}\right).$$

In the opposite limit, $B_{44}^2\ll4|\alpha|EHC_{44}$, the asymptotic values of the excited waves are

$$\omega_{2,3}=\frac{\omega_1}{2}\left(1\pm\sqrt{\frac{(2C_{11}-C_{44})}{C_{11}^2}C_{44}}\right)$$

and contain no explicit dependence on the electric and magnetic fields.

The above process refers to the $GdVO_4$, $DyPO_4$, and $HoPO_4$ compounds (see the literature cited in Ref. 1). One must only bear in mind that the x axis mentioned above must be directed along the same symmetry axis 2 with respect to which the exchange magnetic structure is odd.

5.2. The process related to $\tilde{C}_{166}$

Here we examine the case of an easy-plane antiferromagnet, where the renormalized value of the modulus C_{166} and its dependence on **E** and **H** are especially important. The results refer to the same extent to both $\bar{1}(-)4_z(-)2_d(-)$ and $\bar{1}(-)4_z(+)2_d(-)$ structures, and only for the effective anisotropy constants must we use the appropriate formulas from (19) or (32).

In this case collinear scattering of waves is possible, with $\mathbf{k}_3\downarrow\uparrow\mathbf{k}_2\|\mathbf{k}_1$ (Fig. 2). The first wave with the frequency $\omega_1=k_1\sqrt{C_{11}/\rho}$ is longitudinal, while the generated waves with the frequencies

$$\omega_{23}=k_{2,3}\sqrt{\frac{\tilde{C}_{66}}{\rho}}$$

are transverse waves with polarizations $\mathbf{u}_2\|\mathbf{u}_3\|\mathbf{y}$.

The condition of synchronism (35) in this case leads to the following system of equations:

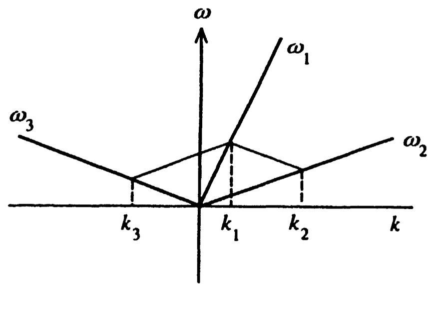

FIG. 2.

$$k_1=k_2-k_3,\quad k_1\sqrt{\frac{C_{11}}{\rho}}=(k_2+k_3)\sqrt{\frac{\tilde{C}_{66}}{\rho}}.$$

Its solution yields the following values for the wave numbers and frequencies of the generated waves:

$$k_{2,3}=\frac{k_1}{2}\left(\sqrt{\frac{C_{11}}{\tilde{C}_{66}}}\pm 1\right),\quad \omega_{2,3}=\frac{1}{2}\,\omega_1\left(1\pm\sqrt{\frac{\tilde{C}_{66}}{C_{11}}}\right).$$

The magnetoelastic and magnetoelectric interactions enter these expressions through the renormalized linear modulus $\tilde{C}_{66}$, which is given by the following expressions:[1]

$$\tilde{C}_{66}=C_{66}-\frac{B_{66}^2}{K_x},$$

where, as noted earlier, K_x must be taken from (19) or (32), depending on whether we are considering the $\bar{1}(-)4_z(-)2_d(-)$ or the $\bar{1}(-)4_z(+)2_d(-)$ structure. The asymptotic behavior of the wave numbers and frequencies at the orientational phase-transition point, where $\tilde{C}_{66}\to 0$, has the following form: $k_{2,3}\to\infty$ and $\omega_{2,3}\to\omega_1/2$.

The above formulas can be applied in the given case to the trirutiles Cr_2WO_6 and $V_2\,WO_6$ $(4_z(-))$ and also to $Cr_2Te\,O_6$ $(4_z(+))$; again see the literature cited in Ref. 1.

6. CONCLUSION

The investigations summarized in the present paper and Ref. 1 show that tetragonal antiferromagnets, to which trirutiles,[5] rare-earth phosphates,[6] and vanadates[7] belong, constitute interesting objects for studying their acoustic properties. Among these compounds there are large groups of antiferromagnets with different exchange magnetic properties (say, the $\bar{1}(-)4_z(+)2_d(-)$ structure in the trirutile Fe_2TeO_6 (Ref. 8) and the $\bar{1}(-)4_z(-)2_d(-)$ structure in Cr_2WO_6 and V_2WO_6 (Ref. 9)) and in different magnetic states ($\mathbf{L}\|4_z$ in $Fe_2\,TeO_6$ and $\mathbf{L}\perp 4_z$ in Cr_2). Some of these have high Néel temperatures Θ_N ($\Theta_N=370\,\mathrm{K}$ for V_2WO_6). Also important is the fact that these antiferromagnets exhibit a magnetoelectric effect. Hence various acoustic phenomena can be observed in them, because they strongly depend on the exchange magnetic structure, the orientation of vector **L** in relation to the crystallographic axes, and the size and direction of the applied external electric and magnetic fields. However, there have been no experimental studies in the acoustics of these compounds. The literature has little or no data on measurement of magnetoelastic constants, static first- and second-order elastic moduli, anisotropy constants, and magnetoelectric constants in such antiferromagnets. For this reason it is difficult to make numerical estimates of the effective anharmonicity with an accuracy greater than that achieved in this paper. Such calculations in the case of antiferromagnets of the easy-plane type require knowing the magnetoelastic constants B_{11}, B_{12}, and B_{66}, the static elastic moduli C_{44}, C_{66}, C_{166}, and C_{144}, and, for the geometry adopted above, the constants $\kappa_\|$ and γ_4 and the anisotropy constants K and K_2. In the case of antiferromagnets of the easy-axis type, for different exchange magnetic structures it is desirable to have the values of the constants B_{11}, B_{12}, B_{44}, and B_{66}, the constant K, the elastic moduli C_{44}, C_{144}, C_{244}, and C_{456}, and also the values of χ, $\kappa_\|$, and γ_4, which together determine the value of α.

The effective anharmonicity of the elastic subsystem in tetragonal antiferromagnets depends not only on the magnetoelastic interaction but also on the magnetoelectric. In easy-axis antiferromagnets with $\mathbf{E}\|\mathbf{H}\|\mathbf{z}$, the contributions to the nonlinear elastic moduli related to the fields **E** and **H** prove to be different for exchange magnetic structures even and odd in relation to the 4_z axis. For the $\bar{1}(-)4_z(+)2_d(-)$ structure only the elastic moduli C_{144}, C_{244}, C_{155}, C_{255}, C_{344}, C_{355}, and C_{456} are effectively renormalized, which is specified by Eqs. (13). Note that this renormalization is present even when $\mathbf{E}=\mathbf{H}=0$. The effective anharmonicity strongly increases near spin-flop transitions, where, for example, $\tilde{C}_{155}$ and $\tilde{C}_{244}$ may reach values $\sim 10^{18}\mathrm{erg/cm^{-3}}$, which exceeds ordinary anharmonicity by a factor of 10^5. However, as Eq. (16) shows, observing such values of $\tilde{C}_{\alpha\beta\gamma}$ requires that a magnetic field $H\sim 10^4\mathrm{Oe}$ be applied to the sample.

For antiferromagnets with an exchange magnetic structure odd with respect to a fourth-order axis, in addition to renormalization of the same moduli as in the previous case there are new dynamical moduli $\tilde{C}_{154}$, $\tilde{C}_{254}$, $\tilde{C}_{345}$, $\tilde{C}_{644}$, and $\tilde{C}_{655}$ (Eq. (14)), which are finite only if the fields **E** and **H** are nonzero. Thus, the emergence of these moduli is caused by the presence of magnetoelectric interaction in the tetragonal antiferromagnets considered here. The greatest values that these moduli may reach are $\sim 10^{18}\,\mathrm{erg/cm^{-3}}$ in the range of reorientation transitions, where the effective anisotropy constants K_x and K_y become minimal.

No marked differences in nonlinear acoustic properties exist in easy-plane antiferromagnets with structures $\bar{1}(-)4_z(+)2_d(-)$ and $\bar{1}(-)4_z(-)2_d(-)$. In both cases the constants renormalized for the geometry of the fields discussed above are C_{166}, C_{266}, C_{144}, C_{244}, C_{344}, and C_{456}. Note that far from orientational transitions, in all the cases considered here, the greatest effective anharmonicity sets in for the moduli C_{166} and C_{266}. For instance, far from transitions the contribution to C_{166} from **E** and **H** is

$$\Delta C_{166}(\mathbf{E},\mathbf{H})\approx\frac{0(B_{11}-B_{12})}{K_2^3}\,\alpha EH.$$

If we assume that $B_{\alpha\beta}\sim 10^6\,\mathrm{erg/cm^{-3}}$, $|K|\sim 10^3\mathrm{erg/cm^{-3}}$, $\alpha\sim 10^{-4}$, $E\sim 10$ esu, and $H\sim 10^4\mathrm{Oe}$, then $\Delta C_{166}(\mathbf{E},\mathbf{H})\sim 10^{12}\mathrm{erg/cm^{-3}}$ and can be of the same order of magnitude as ordinary elastic anharmonicity. The values of C_{166} and C_{266} in the region of spin-reorientation transitions may be of order $10^{18}\mathrm{erg/cm^{-3}}$. A transition of this type is most easily achieved for $K_2<0$ in magnetic fields of about 100 Oe. One more feature of antiferromagnets of the easy-plane type is worth noting. The magnetoelastic and magnetoelectric interactions in this case violate the symmetric relationship $C_{344}=C_{355}$ between the anharmonic moduli, since according to (21) only the nonlinear modulus C_{344} is renormalized. This means that when magnetic ordering emerges, the symmetry of an easy-plane state decreases.

Only in centrally symmetric antiferromagnets has a large number of nonlinear acoustic phenomena been studied experimentally and theoretically. Direct experimental measurements of third-order anharmonic moduli have been done in hemitite,[10] excitation of the second acoustic harmonic in the orthoferrite $TmFeO_3$ has been experimentally observed,[11] and Raman scattering of sound in hemitite has been studied.[12]

In Sec. 5 we demonstrated that there can be a great variety of nonlinear effects in centrally symmetric antiferromagnets. A characteristic feature of such media is that nonlinear processes are affected not only by electric as well as magnetic fields. Hence the experimental study of the various anharmonic effects in such antiferromagnets makes possible a detailed study of the dependence on **E** of the frequency and intensity of the excited waves and of the values of the effective nonlinear elasticity moduli. In view of this the field **E** may serve as one more parameter, in addition to the magnetic field, that can be used to control the processes of nonlinear wave interaction by changing, say, the conditions for synchronism. This fact could play an important role in applications.

Note that we have considered only the features of nonlinear processes that follow directly from the synchronism conditions. Calculating the amplitudes and intensities of the interacting waves requires using the inverse scattering method, following the ideas of Zakharov[13] and allowing for the explicit form of the moduli $\tilde{C}_{\alpha\beta\gamma}$. The explicit form of these moduli must be allowed for because this makes it possible to analyze the magnitudes of the amplitudes and intensities of the excited waves near an orientational phase transition, where the $\tilde{C}_{\alpha\beta\gamma}$ may reach huge values.

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[1)]In contrast to Ref. 1, here the parity of exchange magnetic structures with respect to the symmetry element g is written in the form $g(\pm)$ rather than $g^{\pm}80$ state (see Ref. 3).

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