Softening electromagnetic and sound waves at phase transitions in crystals with linear magnetoelectric effects

1990 J. Phys.: Condens. Matter 2 2579

(http://iopscience.iop.org/0953-8984/2/11/006)

J. Phys.: Condens. Matter 2 (1990) 2579–2601. Printed in the UK

Softening electromagnetic and sound waves at phase transitions in crystals with linear magnetoelectric effects

V G Bar'yakhtar†, I M Vitebskii‡, N M Lavrinenko§ and V L Sobolev‡
† Institute for Metal Physics of Academy of Sciences of the Ukraine, 252650 Kiev, USSR
‡ All Union Scientific Research Institute for Single Crystals, 310141 Kharkov, USSR
§ Donetsk Physico-technical Institute of the Academy of Sciences of the Ukraine, 340114 Donetsk, USSR

Received 15 February 1989, in final form 4 July 1989

Abstract. Whereas the order parameter of a phase transition is transformed either as the components of the electric or magnetic polarisation, or as a linear combination of the deformation tensor components, the velocities of the electromagnetic waves and of sound, which propagate in a definite symmetry direction, become zero when approaching a critical point. A rather large class of such phase transitions of the second kind, the order parameter of which is transformed simultaneously as the electric and magnetic polarisations or as both polarisation and the components of the deformation tensor, is studied in the present paper. In the latter case the bound electromagnetic and sound waves stand for softening hydrodynamic modes. It is shown that at such 'combined' phase transitions qualitatively new effects arise in the behaviour of the softening hydrodynamic waves. The strongest anomalies are observed at magnetoelectric phase transitions.

1. Introduction

In a variety of phase transitions of the second kind the dipole-active and ferroelastic ones occupy a special position. In the first case the order parameter of the phase transition is transformed as some linear combination of the components of the electric p or magnetic m polarisation vectors. In the second one it is transformed as a linear combination of the deformation tensor components $\hat{u}$. The said phase transitions possess some unique properties, which make them different from others.

Every ferroelastic and dipole-active phase transition has a corresponding softening long-wave hydrodynamic mode. For the case of ferroelastics the transverse sound of a definite polarisation, which propagates in some symmetry directions, stands for this mode. At dipole-active transitions the electromagnetic waves play the role of the softening hydrodynamic mode. In both cases the velocity of the softening waves becomes zero, the exception being ferroelastic phase transitions in polarised crystals, where this problem cannot be interpreted unambiguously [1].

The above statement follows from the fact that the velocities of transverse sound v_s and the electromagnetic wave v_c in a crystal are defined by the following expressions:

$$v_s = (\rho s)^{-1/2} \qquad v_c = c(\mu\varepsilon)^{-1/2} \tag{1}$$

where s is the modulus of pliability (the inverse of the shear modulus of a crystal), ρ is the density, c is the light velocity in vacuum, ε and μ are some tensor components of dielectric and magnetic permeabilities. At ferroelastic, ferroelectric or ferromagnetic transitions s, ε or μ values respectively tend to infinity, this being necessary for the velocities v_s or v_c to become zero.

Naturally, such simple considerations are valid only within the hydrodynamic limits, i.e. when

$$\omega \ll \omega_0 \qquad q \ll r_c^{-1} \tag{2}$$

where ω_0 is the lowest of the frequencies characterising the internal degrees of freedom of a crystal and r_c is the correlation length. In this frequency range the effects of spatial and temporal dispersion are not important; s, ε and μ values in (1) coincide with the thermodynamic limits (hydrodynamic principle of local equilibrium). We use these long-wave hydrodynamic limits; all the following considerations will refer to (2).

The described simple pattern of critical behaviour of the softening branches of either sound or electromagnetic waves is related only to those cases where the order parameter of the phase transition is transformed as one of the values p, m or $\hat{u}$. If this is not so, provided the phase transition is simultaneously ferroelastic and dipole-active, the question of the origin of the softening branch (sound or electromagnetic wave?) still remains open. Such 'combined' phase transitions are not uncommon. In particular, the majority of magnetic spin-reoriented transitions are both dipole-active and ferroelastic. Besides, some crystals are known to have both electric and magnetic polarisations involved in the phase transition. In the general case such 'combined' transitions are possible in crystals that permit linear piezoelectric, piezomagnetic or magnetoelectric effects. Consequently, we shall call the mentioned transitions piezoelectric p–$\hat{u}$, piezomagnetic m–$\hat{u}$ and magnetoelectric p–m ones. Obviously, only in the presence of either external magnetic field or spontaneous magnetic ordering may one expect p–m and m–$\hat{u}$ transitions (the time inversion operation $1'$ must be absent in the symmetry group of the initial phase).

Thus, the study of the origin and peculiarities of the critical behaviour of the softening branches of acoustic and electromagnetic waves at ferroelastic and dipole-active transitions in crystals with linear piezoelectric, piezomagnetic or magnetoelectric effects is the main purpose of the present paper. This problem attracts considerable interest first of all due to the originality of the electromagnetic and acoustic properties of such crystals, the most pronounced in the critical region. In such crystals expressions of type (1) for the transverse sound and electromagnetic wave velocities are not applicable even within the hydrodynamic limits q, $\omega \to 0$.

Specifically, in crystals with a linear piezo-effect the excitation dynamics is now described by a common set of bound equations for the electrodynamics and mechanics of the continuous media. And, though the corrections to velocities v_s and v_c from (1) caused by such binding are very small, in a number of cases a qualitative change occurs in the wave propagation pattern, accessible to experimental observation. Finally, in crystals with a linear magnetoelectric effect the velocities of propagation of sound and electromagnetic waves in forward and backward directions may become different. It is these specific effects that may turn out to be most essential and pronounced in the critical region. Their theoretical study is based on a sequential application of group theory methods.

We shall confine our discussion to the softening branches, which almost always are purely transverse. In other words, the alternating electric and magnetic fields, polarisation and displacements due to the propagation of plane electromagnetic and sound waves with small amplitude turn out to be orthogonal to the wavevector q in all cases of interest:

$$E, H, D, B, p, m, u \perp q. \tag{3}$$

The relation (3) makes the analysis of wave equations very simple.

From [2], the set of directions of wavevectors, to which the softening hydrodynamic waves conform, coincides with the set of wavevectors, to which the anomalous critical fluctuations conform. This allows one to use the results from [3] and [4] to search for such directions q. These directions are listed in [5] for ferroelastic transitions in crystals without the magnetoelectric effect.

Dielectric crystals are mainly discussed in the present paper. Effects connected with electrical conductivity may qualitatively change some of the results mentioned below. They are considered in the conclusion.

2. Peculiarities of propagation of magnetic waves at phase transitions in crystals with linear magnetoelectric effect

2.1. *Magnetoelectric transitions and magnetic classes*

Assume there is a bilinear coupling between the order parameter of the phase transition and the components of the vectors p and m. We shall call such transitions magnetoelectric and they are possible only in crystals permitting linear magnetoelectric effect. Of 122 point groups of magnetic symmetry (magnetic classes) such phase transitions are possible in 57; of this number in 43 magnetic classes the said transition conforms to the active irreducible representation. It is these groups that are interesting for further consideration. A lot of specific examples of this kind may be pointed out [6, 7] but here we shall restrict ourselves only to a general consideration.

Piezoelectric and piezomagnetic effects, provided they are available, will be ignored at this stage. This will allow us to study the electromagnetic waves separately from the sound ones. In the next section we shall return to this problem.

In the study of electromagnetic waves in a dielectric medium we assume Maxwell's equations

$$\operatorname{rot} E = -\frac{1}{c}\frac{\partial B}{\partial t} \qquad \operatorname{rot} H = \frac{1}{c}\frac{\partial D}{\partial t} \tag{4}$$

since the remaining two equations

$$\operatorname{div} D = 0 \qquad \operatorname{div} B = 0$$

in case (3) are satisfied automatically.

The relations between the alternating electric and magnetic fields and polarisations, which arise in the electromagnetic wave (all of them are supposed to be weak), have in the most general case the following form [6–9]

$$D = E + 4\pi p = \hat{\varepsilon} E + \hat{\xi} H \tag{5}$$

$$B = H + 4\pi m = \hat{\mu} H + \hat{\xi}^{\mathrm{T}} E \tag{6}$$

where

$$p = \hat{\chi} E + \hat{\psi} H \qquad m = \hat{\kappa} H + \hat{\psi}^{\mathrm{T}} E \tag{7}$$

$$\hat{\varepsilon} = \hat{1} + 4\pi\hat{\chi} \qquad \hat{\mu} = \hat{1} + 4\pi\hat{\kappa} \qquad \hat{\zeta} = 4\pi\hat{\psi}. \tag{8}$$

The tensors $\hat{\zeta}$ and $\hat{\psi}$ in (5)–(8) describe the linear magnetoelectric effect. The sign 'T' in (6) and (7) denotes the transpose operation. In the hydrodynamic limits all the tensors in (5)–(8) are determined under condition of thermodynamic equilibrium, and their form is affected by the magnetic point symmetry of a crystal. The effects of spatial and temporal dispersion are absent in this case. The reverse form of the relations (5)–(8) is also useful:

$$E = \hat{\chi}^{\circ -1}p - \hat{\lambda}m \qquad H = \hat{\kappa}^{\circ -1}m - \hat{\lambda}^{\mathrm{T}}p \tag{9}$$

at the same time

$$\begin{aligned} \hat{\chi}^{\circ} &= \hat{\chi} - \hat{\psi}\hat{\kappa}^{-1}\hat{\psi}^{\mathrm{T}} & \hat{\chi} &= (\hat{\chi}^{\circ -1} - \hat{\lambda}\hat{\kappa}^{\circ}\hat{\lambda}^{\mathrm{T}})^{-1} \\ \hat{\kappa}^{\circ} &= \hat{\kappa} - \hat{\psi}^{\mathrm{T}}\hat{\chi}^{-1}\hat{\psi} & \hat{\kappa} &= (\hat{\kappa}^{\circ -1} - \hat{\lambda}^{\mathrm{T}}\hat{\chi}^{\circ}\hat{\lambda})^{-1} \\ \hat{\lambda} &= [(\hat{\chi}^{-1}\hat{\psi}\hat{\kappa}^{-1})^{-1} - \hat{\psi}^{\mathrm{T}}]^{-1} & \hat{\psi} &= [(\hat{\chi}^{\circ}\hat{\lambda}\hat{\kappa}^{\circ})^{-1} - \hat{\lambda}^{\mathrm{T}}]^{-1}. \end{aligned} \tag{10}$$

In the hydrodynamic limits the relations may be obtained from the equilibrium thermodynamic potential

$$f(E, H) = -\tfrac{1}{2}E \cdot \hat{\chi}E - \tfrac{1}{2}H \cdot \hat{\kappa}H - E \cdot \hat{\psi}H \tag{11}$$

or

$$w(p, m) = \tfrac{1}{2}p \cdot \hat{\chi}^{\circ -1}p + \tfrac{1}{2}m \cdot \hat{\kappa}^{\circ -1}m - p \cdot \hat{\lambda}m. \tag{12}$$

The E and H fields in (11) and (12) are presented as independent parameters, the values of which are determined from Maxwell's equations.

We remind the reader that the vectors E, H, B, D, p and m in the expressions (3)–(12) denote the variable components of the corresponding fields and polarisations related to the electromagnetic wave with small amplitude. Besides, the presence of the constant and homogeneous fields and polarisations is allowed (if it is compatible with the symmetry of the ground state). However, they are insignificant in the present section and we shall not introduce any special designations for them.

The condition of thermodynamic stability for the ground state coincides with the requirement of the positive certainty of the quadratic form (12). Thus, in the stability region of the symmetric phase, the following inequality is satisfied:

$$\Delta = \begin{Vmatrix} \hat{\chi}^{\circ -1} & \hat{\lambda} \\ \hat{\lambda}^{\mathrm{T}} & \hat{\kappa}^{\circ -1} \end{Vmatrix} = \|\hat{\chi}^{-1}\| \cdot \|\hat{\kappa}^{\circ -1}\| = \|\hat{\chi}^{\circ -1}\| \cdot \|\hat{\kappa}^{-1}\| \geqslant 0. \tag{13}$$

On approaching the point of the magnetoelectric phase transition from the side of the symmetric phase:

$$T \to T_{\mathrm{c}} \qquad \Delta \to 0. \tag{14}$$

With this some definite components of the tensors $\hat{\chi}$, $\hat{\kappa}$ and $\hat{\psi}$ (and consequently $\hat{\varepsilon}$, $\hat{\mu}$ and $\hat{\zeta}$) tend to infinity.

Further interpretation is planned as follows: a detailed study will be dedicated to some typical examples and then we shall show the set of magnetoelectric phase transitions to reduce to one of these cases. The most suitable from this point of view are the magnetic classes of the D_{2d} family, which include one non-magnetic class

$$D_{2d} \otimes R \tag{15}$$

Table 1. Transformational properties of t-even $(p, \hat{u})$ and t-odd (m) values relative to transformations from magnetic point groups of the class D_{2d}. Matrices appropriate to the element generators with the unitary part 2_{xy} and σ_x are given in the second column.

	2_{xy}	σ_x		D_{2d} (D_2)	D_{2d} (C_{2v})	D_{2d}	D_{2d} (S_4)
A_1	1	1					m_z
A_2	$\bar{1}$	$\bar{1}$				m_z	
B_1	1	$\bar{1}$	u_{xy}		m_z		
B_2	$\bar{1}$	1	$p_z, u_{xx} - u_{yy}$	m_z			
E	$\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$	$\begin{pmatrix} \bar{1} & 0 \\ 0 & 1 \end{pmatrix}$	$\left.\begin{matrix} p_x \\ p_y \end{matrix}\right\}\ \left.\begin{matrix} u_{xz} \\ -u_{yz} \end{matrix}\right\}$	$\left.\begin{matrix} m_x \\ m_y \end{matrix}\right\}$	$\left.\begin{matrix} m_y \\ -m_x \end{matrix}\right\}$	$\left.\begin{matrix} m_y \\ m_x \end{matrix}\right\}$	$\left.\begin{matrix} m_x \\ -m_y \end{matrix}\right\}$

(where R is the group, consisting of 1 and 1′ elements) and four classes with the magnetic order:

$$(1)\ D_{2d}(D_2) \qquad (2)\ D_{2d}(C_{2v}) \qquad (3)\ D_{2d} \qquad (4)\ D_{2d}(S_4). \tag{16}$$

Note that the first three magnetic classes from (16) are purely antiferromagnetic and the last, the fourth, permits the existence of the magnetic moment along the tetragonal axis.

The distribution of the Cartesian components of p, m and $\hat{u}$ values along the irreducible representations of the magnetic point groups of the D_{2d} family is given in table 1. The numerical order of the irreducible representation is given in the first column. The representation matrices for the element generators with unitary part 2_{xy} and σ_x are given in the second column. In the third and following columns the distribution of t-even and t-odd values along the irreducible representations of different magnetic point groups is given respectively. The transformation properties of t-even values (p and $\hat{u}$) are equal for all the magnetic groups and that is why they are generalised for all the groups in the third column of table 1.

It turns out that all the qualitatively different types of behaviour of the softening sound and electromagnetic waves at dipole-active and ferroelastic phase transitions are realised at the transitions along the two-dimensional irreducible representation E of every point group (15, 16). Therefore, all the examples considered in the following sections apply just to this irreducible representation. The examples for other magnetic classes are studied in [2] and are given in table 2.

In all the cases under investigation the direction $q \| z$ is appropriate to the softening branches of the spectrum; consequently only this direction of propagation will be considered hereafter†.

According to (3) only x and y components of the field, polarisation and displacement vectors will be other than zero in the softening waves. The corresponding contributions to the expressions for $f(E, H)$ and $w(p, m)$ from (11) and (12) (the elastic degrees of freedom are ignored in the present section) have the form

$$f(E, H) = -\tfrac{1}{2}\chi(E_x^2 + E_y^2) - \tfrac{1}{2}\kappa(H_x^2 + H_y^2) - \psi_{\alpha\beta} E_\alpha H_\beta \tag{17}$$

$$w(p, m) = \frac{1}{2\chi^\circ}(p_x^2 + p_y^2) + \frac{1}{2\kappa^\circ}(m_x^2 + m_y^2) - \lambda_{\alpha\beta} p_\alpha m_\beta \tag{18}$$

where $\alpha, \beta = x, y$. The form of $\hat{\psi}$ and $\hat{\lambda}$ tensors of the magnetoelectric effect is different

† In consequence of [4] and table 2, [110] and [1$\bar{1}$0] directions also conform to the softening waves for D_{2d}, $D_{2d} \otimes R$ and $D_{2d}(D_2)$ groups.

Table 2. Magnetoelectric transitions at which the vector components p and m arising in a dissymmetric phase are non-collinear. The last two columns refer to the phase transitions of the p–m–$\hat{u}$ type. The following designations are used in part (c): a and b are two mutually normal directions in the xy plane; $q_{1,2} \parallel [\pm 1, 1, 0]$.

(a)

	p_α	m_α	q_α	v°_α	$u_{\alpha\beta}$	q^*_α
C_i (C_1)	p_x, p_y, p_z	m_x, m_y, m_z	$q_{x,y,z}$	v_x, v_y, v_z		
C_2	p_x, p_y	m_x, m_y	q_z	v_z	u_{xz}, u_{yz}	q_z
C_s	p_z	m_x, m_y	$q_{x,y}$	v_x, v_y	u_{xz}, u_{yz}	—
C_2 (C_1)	p_x, p_y	m_z	$q_{x,y}$	v_x, v_y	u_{xz}, u_{yz}	—
C_{2h} (C_2)	p_x, p_y	m_x, m_y	q_z	v_z		
C_{2h} (C_s)	p_z	m_x, m_y	$q_{x,y}$	v_x, v_y		
	p_x, p_y	m_z	$q_{x,y}$			
C_{2v} (C_s)	p_x	m_y	q_z	v_z	u_{xy}	—
C_{2v}	p_x	m_y	q_z	v_z	u_{xz}	q_z
	p_y	m_x	q_z		u_{yz}	q_z
D_2 (C_2)	p_x	m_y	q_z	v_z	u_{yz}	q_z
	p_y	m_x	q_z		u_{xz}	q_z
D_{2h}(C_{2v})	p_x	m_y	q_z	v_z		
	p_y	m_x	q_z			

(b)

	p_α	m_α	q_α	v°_α	$u_{\alpha\beta}$	q^*_α
S_4(C_2)	$\begin{cases} p_+ \\ p_- \end{cases}$	m_+ m_-	q_z	v_z	u_{z-} u_{z+}	q_z
S_6(C_3), C_{4h}(C_4), C_{6h}(C_6)	$\begin{cases} p_+ \\ p_- \end{cases}$	m_+ m_-	q_z	v_z		
D_{3d}(C_{3v}), D_{4h}(C_{4v}), D_{6h}(C_{6v})	$\begin{cases} p_x \\ p_y \end{cases}$	m_y $-m_x$	q_z	v_z		
C_{4v}, C_{6v}	$\begin{cases} p_x \\ p_y \end{cases}$	m_y $-m_x$	q_z	v_z	u_{xz} u_{yz}	q_z
D_{2d}(C_{2v})	$\begin{cases} p_x \\ p_y \end{cases}$	m_y $-m_x$	q_z	v_z	u_{xz} $-u_{yz}$	q_z

(c)

	p_α	m_α	q_α	v°_α	$u_{\alpha\beta}$	q^*_α
S_4	$\begin{cases} p_+ \\ p_- \end{cases}$	m_- m_+	$q \perp a, q \perp b$	—	u_{z-} u_{z+}	q_z
C_{4h}(S_4)	$\begin{cases} p_+ \\ p_- \end{cases}$	m_- m_+	$q \perp a, q \perp b$	—		
D_{4h}(D_{2d})	$\begin{cases} p_x \\ p_y \end{cases}$	m_y m_x	$q \perp q_1, q \perp q_2$	—		
D_{2d}	$\begin{cases} p_x \\ p_y \end{cases}$	m_y m_x	$q \perp q_1, q \perp q_2$	—	u_{xz} $-u_{yz}$	$q_z, q_{1,2}$
D_{2d}(S_4)	$\begin{cases} p_x \\ p_y \end{cases}$	m_x $-m_y$	$q \perp x, q \perp y$	—	u_{xz} $-u_{yz}$	q_z
C_{4v}(C_{2v})	$\begin{cases} p_x \\ p_y \end{cases}$	m_y m_x	$q \perp q_1, q \perp q_2$	—	u_{xz} u_{yz}	q_z

for every magnetic class (16). From table 1 we have four cases:

Case 1. Magnetic class $D_{2d}(D_2)$

$$\psi_{\alpha\beta}E_\alpha H_\beta = \psi(E_xH_x + E_yH_y) \qquad \lambda_{\alpha\beta}p_\alpha m_\beta = \lambda(p_xm_x + p_ym_y). \tag{19}$$

This case is typical for the state with $p \| m$ to arise as a result of the magnetoelectric phase transitions.

Case 2. Magnetic class $D_{2d}(C_{2v})$

$$\psi_{\alpha\beta}E_\alpha H_\beta = \psi(E_xH_y - E_yH_x) \qquad \lambda_{\alpha\beta}p_\alpha m_\beta = \lambda(p_xm_y - p_ym_x). \tag{20}$$

The fact that p and m are not parallel in the dissymmetric phase is most essential here. In this case $p \perp m$.

Case 3. Magnetic class D_{2d}

$$\psi_{\alpha\beta}E_\alpha H_\beta = \psi(E_xH_y + E_yH_x) \qquad \lambda_{\alpha\beta}p_\alpha m_\beta = \lambda(p_xm_y + p_ym_x). \tag{21}$$

The structure of the combined invariant in (21) is such that mutual orientation of p and m vectors in the xy plane is not fixed. Actually

$$\lambda(p_xm_y + p_ym_x) = \lambda pm \sin(\varphi_p + \varphi_m)$$

and therefore the value of the angle between vectors p and m (the difference $(\varphi_p - \varphi_m)$) does not affect any summand in (18).

Case 4. Magnetic class $D_{2d}(S_4)$

$$\psi_{\alpha\beta}E_\alpha H_\beta = \psi(E_xH_x - E_yH_y) \qquad \lambda_{\alpha\beta}p_\alpha m_\beta = \lambda(p_xm_x - p_ym_y). \tag{22}$$

Cases 3 and 4 are equivalent, accurate to a rotation of the coordinate system by the angle $\pi/4$ in the xy plane.

The stability region of the symmetric phase in the cases (19)–(22) is defined by an inequality

$$\xi = 1 - \lambda^2\chi^\circ\kappa^\circ = 1 - \psi^2/\kappa\chi \geqslant 0 \tag{23}$$

which will be assumed to be satisfied. The values

$$\begin{array}{lll} \chi = \chi^\circ/\xi & \kappa = \kappa^\circ/\xi & \psi = \lambda\kappa^\circ\chi^\circ/\xi \\ \varepsilon = 1 + 4\pi\chi & \mu = 1 + 4\pi\kappa & \zeta = 4\pi\psi \end{array} \tag{24}$$

coincide with the components of the corresponding tensors from (5)–(11), which diverge at the critical point: at $T \to T_c$

$$\xi \to 0 \qquad \chi, \kappa, \varepsilon, \mu, \psi, \zeta \to \infty. \tag{25}$$

After all these preliminary remarks, we turn back to the Maxwell equations and consider them individually for cases 1, 2 and 3. It will be shown below that the variety of magnetoelectric phase transitions can be reduced to one of these three cases.

With allowance for the relations (5) and (6) we obtain from (4) for the plane electromagnetic wave

$$q \times E = \frac{\omega}{c}(\hat{\mu}H + \hat{\zeta}^{\mathrm{T}}E) \qquad q \times H = -\frac{\omega}{c}(\hat{\varepsilon}E + \hat{\zeta}H). \tag{26}$$

We shall consider only $q \| z$, which is appropriate to the softening modes of the electromagnetic waves. The expressions (17)–(24) then fully define the relations necessary for the solution of the set of linear equations (26).

2.2. Maxwell equations in various cases

2.2.1. Case 1. Magnetic class $D_{2d}(D_2)$. In this case the set of linear equations (26) with (17) and (19) allowed for is reduced to the following form

$$\begin{aligned} &(1 + i\zeta v/c)E_+ + i\mu(v/c)H_+ = 0 \\ &-i\varepsilon(v/c)E_+ + (1 - i\zeta v/c)H_+ = 0 \end{aligned} \tag{27}$$

$$\begin{aligned} &(1 - i\zeta v/c)E_- - i\mu(v/c)H_- = 0 \\ &i\varepsilon(v/c)E_- + (1 + i\zeta v/c)H_- = 0 \end{aligned} \tag{28}$$

where

$$v = \omega/q \qquad E_\pm = E_x \pm iE_y \qquad H_\pm = H_x \pm iH_y. \tag{29}$$

The sets of equations (27) and (28) are equivalent, accurate to complex conjugation.

The velocities of propagation of electromagnetic waves in $+z$ and $-z$ directions are equal respectively to

$$v/c = \pm(\varepsilon\mu - \zeta^2)^{-1/2} = \pm\, \xi^{1/2}(\varepsilon^\circ\mu^\circ - 1 + \xi)^{-1/2}. \tag{30}$$

Here, in consequence of (8) and (24),

$$\varepsilon^\circ = 1 + 4\pi\chi^\circ \qquad \mu^\circ = 1 + 4\pi\kappa^\circ.$$

On approaching the point of the magnetoelectric transition from the side of the symmetric phase we have: at $T \to T_c$

$$v/c \simeq \pm\xi^{1/2}(\varepsilon^\circ\mu^\circ - 1)^{-1/2} \to 0 \tag{31}$$

$$\kappa^{\circ 1/2} p_\pm \simeq (\operatorname{sgn}\lambda)\chi^{\circ 1/2} m_\pm \tag{32}$$

$$\chi^{\circ 1/2} E_\pm \simeq -(\operatorname{sgn}\lambda)\kappa^{\circ 1/2} H_\pm \simeq -i\xi^{1/2} \frac{4\pi\chi^{\circ 1/2}}{(\varepsilon^\circ\mu^\circ - 1)^{1/2}} m_\pm \ll m_\pm, p_\pm \tag{33}$$

where $\operatorname{sgn}\lambda = \lambda/|\lambda|$.

Let us note that at $T \to T_c$ magnetic and electric polarisations both in the softening electromagnetic wave and in quasi-stationary fluctuation wave [2, 4] are defined by one and the same expression. This absolutely general circumstance results in the coincidence of the set of wavevectors, to which the abnormal fluctuations conform, and for which the softening hydrodynamic modes exist.

Thus, in the hydrodynamic region (2) the velocity of the softening electromagnetic waves tends to zero as $\xi^{1/2}$, which is in principle analogous to the case of either purely ferromagnetic or purely ferroelectric phase transition (see the expression (1) for v_c). The peculiar behaviour of the polarisation of the softening modes due to the presence of the linear magnetoelectric effect, which is defined by the invariant of the form (19), is the only difference.

2.2.2. Case 2. Magnetic class $D_{2d}(C_{2v})$. In this case at $q \| z$ the set of equations (26) with allowance for (17) and (20) also split into independent blocks, describing electromagnetic waves of different polarisation:

$$\begin{aligned}&(1)\ E\|p\|x \qquad H\|m\|y\\&(2)\ E\|p\|y \qquad H\|m\|x.\end{aligned}\tag{34}$$

For the first and second polarisations we have respectively

$$(1)\ \begin{aligned}&(1-\zeta v/c)E_x-\mu(v/c)H_y=0\\&-\varepsilon(v/c)E_x+(1-\zeta v/c)H_y=0\end{aligned}\tag{35}$$

$$(2)\ \begin{aligned}&(1-\zeta v/c)E_y+\mu(v/c)H_x=0\\&\varepsilon(v/c)E_y+(1-\zeta v/c)H_x=0.\end{aligned}\tag{36}$$

The sets of equations (35) and (36) are equivalent, accurate to a rotation by an angle of $\pi/2$ round the z axis, which is the result of the isotropy of the problem in the xy plane (the said isotropy occurs only in cases 1 and 2, which can be seen from the expressions (17)–(22)).

The velocities of propagation of the electromagnetic waves of both polarisations in $+z$ and $-z$ directions are respectively

$$v/c=[\zeta\pm(\varepsilon\mu)^{1/2}]^{-1}\tag{37}$$

or in an equivalent form (in accordance with (24))

$$v/c=\xi\{4\pi(\operatorname{sgn}\lambda)[(1-\xi)\chi^\circ\kappa^\circ]^{1/2}\pm[(\xi+4\pi\chi^\circ)(\xi+4\pi\kappa^\circ)]^{1/2}\}^{-1}.\tag{38}$$

On approaching the point of phase transition we have from (38) at $T\to T_c$

$$v/c\simeq(\operatorname{sgn}\lambda)\begin{cases}\dfrac{\xi}{8\pi(\kappa^\circ\chi^\circ)^{1/2}}=\dfrac{1}{8\pi(\chi\kappa)^{1/2}}\to 0 & (39)\\[2ex] -\dfrac{8\pi(\kappa^\circ\chi^\circ)^{1/2}}{\varepsilon^\circ\mu^\circ-1}\neq 0. & (40)\end{cases}$$

The structure of the softening mode (39) for the polarisation 1 is defined by the expressions

$$\kappa^{\circ 1/2}p_x\simeq(\operatorname{sgn}\lambda)\chi^{\circ 1/2}m_y\tag{41}$$

$$\chi^{\circ 1/2}E_x\simeq(\operatorname{sgn}\lambda)\kappa^{\circ 1/2}H_y\simeq\frac{\xi}{2\chi^{\circ 1/2}}p_x\ll p_x,m_y\tag{42}$$

which correspond exactly to the structure of the quasi-stationary fluctuation wave (see the note after formula (33)). For the polarisation 2 we have similar expressions, differing from (41) and (42) by a rotation round the z axis by an angle of $\pi/2$.

Undoubtedly, the violation of the reciprocity law, i.e. the velocities of the electromagnetic waves in the forward and backward directions are different in magnitude, is the most peculiar feature in the situation described above. This difference is most pronounced at the critical point of the magnetoelectric phase transition, where the velocity of the electromagnetic wave becomes zero only for one of the possible directions of propagation ($+z$ or $-z$), depending on the sign of λ (see formulae (39) and (40)).

It is also important that the critical anomaly in the velocity of propagation of the electromagnetic waves is here much stronger than in all previously described cases. Indeed, in case 1 and also at dipole-active phase transitions in crystals, having no linear magnetoelectric effect, the velocity of the electromagnetic waves at $T \to T_c$ tends to zero as $\xi^{1/2}$. But in case 2, according to (39) the velocity of the softening wave is proportional to ξ. Note that with the disregard of the fluctuation effects, $\xi \propto |T - T_c|$.

2.2.3. Case 3. Magnetic class D_{2d}. At $q \| z$ for the waves with polarisations 1 and 2 from (34) we have, according to (26), (17) and (21),

$$(1)\quad \begin{aligned} &(1 - \zeta v/c)E_x - \mu(v/c)H_y = 0 \\ &-\varepsilon(v/c)E_x + (1 - \zeta v/c)H_y = 0 \end{aligned} \tag{43}$$

$$(2)\quad \begin{aligned} &(1 + \zeta v/c)E_y + \mu(v/c)H_x = 0 \\ &\varepsilon(v/c)E_y + (1 + \zeta v/c)H_x = 0. \end{aligned} \tag{44}$$

The velocities of propagation for the electromagnetic waves with polarisations 1 and 2 in the $\pm z$ directions are

$$(1)\ v/c = [\zeta \pm (\varepsilon\mu)^{1/2}]^{-1} \tag{45}$$

$$(2)\ v/c = [-\zeta \pm (\varepsilon\mu)^{1/2}]^{-1} \tag{46}$$

or in other notation (see (24))

$$(1)\ v/c = \xi\{4\pi(\operatorname{sgn}\lambda)[(1-\xi)\chi^\circ\kappa^\circ]^{1/2} \pm [(\xi + 4\pi\chi^\circ)(\xi + 4\pi\kappa^\circ)]^{1/2}\}^{-1} \tag{47}$$

$$(2)\ v/c = \xi\{-4\pi(\operatorname{sgn}\lambda)[(1-\xi)\chi^\circ\kappa^\circ]^{1/2} \pm [(\xi + 4\pi\chi^\circ)(\xi + 4\pi\kappa^\circ)]^{1/2}\}^{-1}. \tag{48}$$

The $\pm$ signs in (45)–(48), as in the expressions (30), (31), (37) and (38), refer to the electromagnetic waves propagating in two opposite directions along the z axis.

Finally, on approaching the point of phase transition we obtain for the polarisations 1 and 2: at $T \to T_c$

$$(1)\ v/c \simeq (\operatorname{sgn}\lambda)\begin{cases} \dfrac{\xi}{8\pi(\chi^\circ\kappa^\circ)^{1/2}} = \dfrac{1}{8\pi(\chi\kappa)^{1/2}} \to 0 & (49) \\[2ex] -\dfrac{8\pi(\chi^\circ\kappa^\circ)^{1/2}}{\varepsilon^\circ\mu^\circ - 1} \neq 0 & (50) \end{cases}$$

$$(2)\ v/c \simeq -(\operatorname{sgn}\lambda)\begin{cases} \dfrac{\xi}{8\pi(\chi^\circ\kappa^\circ)^{1/2}} = \dfrac{1}{8\pi(\chi\kappa)^{1/2}} \to 0 & (51) \\[2ex] -\dfrac{8\pi(\kappa^\circ\chi^\circ)^{1/2}}{\varepsilon^\circ\mu^\circ - 1} \neq 0. & (52) \end{cases}$$

The structure of the softening modes is defined by the expressions

$$(1)\ \kappa^{\circ 1/2} p_x \simeq (\operatorname{sgn}\lambda)\chi^{\circ 1/2} m_y \qquad (\text{mode (49)}) \tag{53}$$

$$(2)\ \kappa^{\circ 1/2} p_y \simeq -(\operatorname{sgn}\lambda)\chi^{\circ 1/2} m_x \qquad (\text{mode (51)}). \tag{54}$$

Let us note that every formula concerning polarisation 1 for case 3 coincides with the corresponding formula for case 2 (expressions (35) and (37)–(42)). As for the electromagnetic wave with polarisation 2, the expressions describing it differ from (36)–(40) only in the sign of $\lambda(\zeta)$.

Thus, in case 3, for each of the polarisations 1 or 2 from (34) the velocities of propagation of the electromagnetic waves in the $+z$ and $-z$ directions differ in magnitude, as in case 2. However, the velocity of the wave with polarisation 1 (2) and the velocity of the wave with polarisation 2 (1), propagating in the opposite direction, are equal in magnitude. At $T \rightarrow T_c$ the velocity of the softening electromagnetic waves is linearly decreased along ξ as in case 2.

It should be noted that in case 3 some other directions of propagation conform to the softening waves (see [4] and table 2). However, at $q \neq q_z$ the velocity of the softening waves decreases essentially more slowly—as $\xi^{1/2}$. A more detailed analysis is given in [2].

Since this problem is of special importance we shall now thoroughly consider the symmetry aspects of the above-described reciprocity relations.

2.3. *Symmetry aspects of reciprocity relations*

The spectrum of the arbitrary linear excitations (plane waves) consists of a set of frequencies $\{\omega_n(q)\}$ where n is the number of the branch of the spectrum. We assume that the reciprocity law is satisfied in the wide sense provided the sets of frequencies $\{\omega_n(q)\}$ and $\{\omega_n(-q)\}$ coincide. The numerical order of the frequencies in these sets may be different, e.g. $\omega_1(q) = \omega_2(-q)$, etc.

To satisfy the reciprocity principle it is necessary and sufficient for the symmetry group of the crystal G to contain an operation that would change the sign of the t-odd polar vector v, parallel to q

$$\hat{g}v = -v \qquad \hat{g} \in G \qquad v \| q. \tag{55}$$

If group G contains the inversion $\bar{1}$ or the time inversion operation $1'$, then the condition (55) is satisfied at any q and the sets of frequencies $\{\omega_n(-q)\}$ and $\{\omega_n(q)\}$ coincide.

Such fulfilment of criterion (55) means that the reciprocity principle (in the wide sense) is satisfied for any plane linear waves in the given medium for the direction of propagation pointed out in (55). Let us consider just the electromagnetic waves. Here we can find such an effect where the violation of the reciprocity principle, unless the criterion (55) is satisfied, can only be the result of spatial dispersion and, therefore, falls outside of the hydrodynamic consideration. In the hydrodynamic approximation the reciprocity principle is violated only in crystals with linear magnetoelectric effect. Indeed, at $\xi = 0$ equations (26) are known to be invariant relative to the substitution

$$q \rightarrow -q \qquad \omega \rightarrow \omega \qquad H \rightarrow -H \qquad E \rightarrow E. \tag{56}$$

Analysis of all possible magnetoelectric phase transitions shows that for the softening branches of the spectrum of electromagnetic waves the reciprocity principle in the hydrodynamic limit is satisfied when a condition weaker than in (55) is obeyed:

$$v^\circ \equiv \sum_{\hat{g} \in G} \hat{g}v = 0 \qquad v \| q. \tag{57}$$

The vector v° from (57) is known to be an invariant of the group G. Therefore, the invariance of maybe one t-odd polar vector v° is a sufficient condition for the violation of the reciprocity principle for the electromagnetic waves in the hydrodynamic limit.

Simple comments concerning the idea of criterion (57) are as follows: Given $v \| z$, the invariance v_z relative to the operation from G means also the invariance of the bilinear form

$$p_x m_y - p_y m_x \tag{58}$$

since the transformational properties of v_z and the values (58) coincide. But from the analysis of case 2, it can be seen that it is just the invariance of type (58) in the expression for $w(p, m)$ that is responsible for the violation of the reciprocity principle.

The possibility of violation of the reciprocity principle for the electromagnetic waves in the hydrodynamic case is pointed out in [10]. The situation there conforms to the magnetic classes D_2 or $D_{2h}(D_2)$. In this case the velocities of the electromagnetic waves do not coincide for the forward and backward directions, only when the wavevector has all three non-zero projections onto the rhombic axes (note that the criterion (55) is satisfied provided $q_x q_y q_z = 0$). This case is not of interest to us, since the directions normal to one of the second-order axes conform to the softening branches in the said magnetic classes. More detailed information on the aspects of the electrodynamics in magnetoelectric media is given in [6, 7].

The violation of the reciprocity principle in the wide sense for the softening electromagnetic waves—both conditions (55) and (57) at $q \| z$ are not obeyed with all the consequences ensuing therefrom—is the most significant feature in the above considered case 2.

A quite different situation is realised in case 3 (magnetic class D_{2d}). Both criteria (55) and (57) are satisfied here (at $q \| z$), and therefore the reciprocity principle in the wide sense holds true even outside the limits of the hydrodynamic approximation. Nevertheless, for any two polarisations (34), the velocities of propagation of the electromagnetic wave in the $+z$ and $-z$ directions differ in magnitude (see formulae (45) and (46)). In this case the violation of the reciprocity principle is to be understood in the narrow sense. With the relations (56) allowed for, it is easy to understand that in the hydrodynamic limit the said violation even in the narrow sense is possible only in crystals with linear magnetoelectric effect.

2.4. Critical behaviour in various cases

The relations described above present a reasonable basis for the classification of possible types of behaviour of the softening electromagnetic waves at magnetoelectric phase transitions. In a crystal with linear magnetoelectric effect at any set directions of q, four electromagnetic waves, which differ in polarisation and direction of propagation, should be considered. The velocities of propagation of these waves will be denoted respectively

$$v_+(1) \qquad v_-(1) \qquad v_+(2) \qquad v_-(2) \tag{59}$$

where the + and − signs denote two opposite directions of propagation and the numbers 1 and 2 denote the polarisation of the waves.

At the critical point of the magnetoelectric phase transition with definite directions of q, some of the values (59) become zero. The analysis carried out for crystals with different magnetic symmetry shows that three types of critical behaviour for the softening branches of the spectrum of the electromagnetic waves are possible in accordance with those three examples discussed above.

2.4.1. Case 1. At $T \to T_c$

$$\text{(a) } v_+(1) = -v_-(1) = v_+(2) = -v_-(2) \sim \xi^{1/2} \to 0 \tag{60}$$

$$\text{(b) } v_+(1) = -v_-(1) \sim \xi^{1/2} \to 0 \qquad v_+(2) = -v_-(2) \neq 0. \tag{61}$$

In case 1 the reciprocity principle is valid and the critical behaviour of the velocity of the softening electromagnetic waves is analogous to the case of the dipole-active phase transitions in crystals without any linear magnetoelectric effect. Only a specific behaviour of the polarisation of the softening modes testifies to such an effect. Case 1 is realised in those crystals where electric and magnetic polarisations are related as

$$\lambda_{\alpha\beta} p_\alpha m_\beta = \lambda p_\gamma m_\gamma. \tag{62}$$

The Cartesian index runs over values 1, 2 or 3, depending upon the dimensionality of the irreducible representation responsible for the phase transition. And if the direction q coincides with a symmetry axis above second order, both polarisations are equivalent (case (60)); otherwise the electromagnetic wave of only one of the polarisations is softened (case (61)).

2.4.2. *Case 2. At* $T \to T_c$

$$\text{(a) } v_+(1) = v_+(2) \sim \xi \to 0 \qquad v_-(1) = v_-(2) \neq 0 \tag{63}$$

$$\text{(b) } v_+(1) \sim \xi \to 0 \qquad v_-(1), v_+(2), v_-(2) \neq 0. \tag{64}$$

Similarly to case 1, the expression (a) corresponds to the situation where the direction of wave propagation q coincides with a symmetry axis of higher than second order.

The violation of the reciprocity principle in the wide sense—a discrepancy in the velocities of the electromagnetic waves that propagate in the forward and backward directions—is the most characteristic feature of case 2. Note that at the critical point this discrepancy is most pronounced. Besides, the critical anomaly in the velocity of the softening waves is much more pronounced in case 2 than in case 1 and in crystals without linear magnetoelectric effect, i.e. on approaching T_c the velocity of the wave is decreased linearly along ξ, but not as $\xi^{1/2}$ in case 1. The list of magnetic classes related to case 2 is given in tables 2(*a*) and (*b*).

2.4.3. *Case 3. At* $T \to T_c$, $q \| z$

$$v_+(1) = -v_-(2) \sim \xi \to 0 \qquad v_-(1) = -v_+(2) \neq 0. \tag{65}$$

In this case the reciprocity principle is satisfied in the wide sense. However, for each of two polarisations the velocity of the electromagnetic waves for both directions is different, i.e. in the narrow sense the reciprocity principle is violated. Case 3 is realised only at the magnetoelectric phase transitions along the two-dimensional irreducible and physically irreducible representations in tetragonal crystals. The list of the corresponding magnetic classes is given in table 2(*c*).

Let us note that, in every magnetic class related to case 3, along with the z axis there are two more mutually normal planes to which the softening branches of the electromagnetic waves spectrum conform. However, if q is not parallel to z, the critical anomalies in the velocities of propagation of the corresponding waves are weaker ($v \sim \xi^{1/2}$).

2.5. *Summary*

A more detailed description of the results concerning different magnetoelectric phase transitions is given below.

Of 43 magnetic classes, having common active irreducible representations for the vector components p and m, in 17 the corresponding combined invariants have the structure (62) and are related to a less interesting case 1. The results for the remaining 26 point groups of magnetic symmetry are shown in table 2; note that case 2 is appropriate to tables 2(*a*) and (*b*), and case 3 to table 2(*c*).

The list of the corresponding 26 magnetic classes is in the first column of table 2. The basis functions constructed of the vector components p and m and transforming along the irreducible representations responsible for the magnetoelectric phase transition are given in the second and third columns.

It is the magnetic classes containing only one-dimensional representations that are given in table 2(*a*) (there are two irreducible representations of the type p–m in every group $C_{2h}(C_s)$, C_{2v}, $D_2(C_2)$, $D_{2h}(C_{2v})$). Two-dimensional irreducible and physically irreducible representations are given in tables 2(*b*) and (*c*); the corresponding basis functions are also shown. In consequence of [9] we shall consider the operation of time inversion 1′ to change the sign of the t-odd values, however still being linear† (not connected with complex conjugation). Bilinear invariants, which conform to the physically irreducible representations, have the structure

$$p_+m_+^* = p_+m_- \rightarrow \begin{cases} p_xm_x + p_ym_y \\ p_xm_y - p_ym_x \end{cases} \tag{66}$$

$$p_+m_-^* = p_+m_+ \rightarrow \begin{cases} p_xm_x - p_ym_y \\ p_xm_y + p_ym_x. \end{cases} \tag{67}$$

The invariants of type (66) and (67) arise respectively for the cases in tables 2(*b*) and (*c*). The directions of the wavevector to which the softening branches of the electromagnetic waves spectrum correspond are pointed out in the fourth column of table 2. In the cases from tables 2(*a*) and (*b*) only the direction of the wavevector is appropriate to the softening waves and in the cases from table 2(*c*) the corresponding wavevectors lie in the mutually normal planes having the crystallographic axis z.

The invariant components of the polar t-odd vector v from (57) are given in the fifth column. For the cases in table 2(*c*) $v^\circ = 0$ at any v.

Some of the magnetoelectric phase transitions are at the same time proper ferroelastic ones. Here they are presented by p–m–$\hat{u}$ transitions. Tensor components of the deformations that are transformed as the order parameter of the phase transition are given in the sixth column for such phase transitions. Striction interactions at phase transitions result in the decrease of the number of directions of wavevectors to which the softening hydrodynamic waves are appropriate. In accordance with this, the directions of propagation of the softening waves, still taking into account all three long-range interactions, are given in the last column of table 2. A dash in the last column means that the softening hydrodynamic modes (bound sound and electromagnetic waves) are absent in this case‡. A detailed description of these items will be quoted in the following section.

† Such an approach is justified only for the study of the static and quasi-static processes, i.e. in the limits of applicability of the hydrodynamic principle of local equilibrium.

‡ In the same cases, a simultaneous account of three long-range interactions—electric dipole, magnetic dipole and striction interactions—leads to a complete suppression of anomalous critical fluctuations at the point of phase transition of the type p–m–$\hat{u}$ [4]. Let us note that such a complete striction suppression of the critical fluctuations is also possible at a phase transition without any change of the symmetry in the vicinity of the critical point on the line of the phase transition of the first kind [11, 12].

3. Bound electromagnetic and sound waves in crystals with linear piezo-effect

Maxwell's equations (4) and Newton's equation are assumed in dynamic form in this case. Newton's equation is taken as

$$\rho \ddot{u}_\alpha = \frac{\partial}{\partial x_\beta} \sigma_{\beta\alpha} \tag{68}$$

where $\hat{\sigma}$ is the stress tensor.

For plane waves, instead of (4) and (68) we have

$$q \times E = \frac{\omega}{c} B \qquad q \times H = -\frac{\omega}{c} D$$
$$\rho\omega^2 u_{\alpha,\beta} = q_\beta q_\gamma \sigma_{\gamma\alpha}. \tag{69}$$

For a simplification both parts of equation (68) are coordinate differentiated. Taking into account the possibility of the existence of linear piezoelectric and piezomagnetic effects, we have instead of (5)–(7) the following relations:

$$\begin{aligned} D_\alpha &= \varepsilon_{\alpha\beta} E_\beta + \zeta_{\alpha,\beta} H_\beta + \zeta^{(1)}_{\alpha,\beta\gamma} \sigma_{\beta\gamma} \\ B_\alpha &= \zeta_{\beta,\alpha} E_\beta + \mu_{\alpha\beta} H_\beta + \zeta^{(2)}_{\alpha,\beta\gamma} \sigma_{\beta\gamma} \\ u_{\alpha\beta} &= \psi^{(1)}_{\gamma,\alpha\beta} E_\gamma + \psi^{(2)}_{\gamma,\alpha\beta} H_\gamma + s_{\alpha\beta,\gamma\delta} \sigma_{\gamma\delta} \end{aligned} \tag{70}$$

$$\begin{aligned} p_\alpha &= \chi_{\alpha\beta} E_\beta + \psi_{\alpha,\beta} H_\beta + \psi^{(1)}_{\alpha,\beta\gamma} \sigma_{\beta\gamma} \\ m_\alpha &= \psi_{\beta,\alpha} E_\beta + \kappa_{\alpha\beta} H_\beta + \psi^{(2)}_{\alpha,\beta\gamma} \sigma_{\beta\gamma}. \end{aligned} \tag{71}$$

The tensors $\hat{\zeta}^{(1)} = 4\pi\hat{\psi}^{(1)}$ and $\hat{\zeta}^{(2)} = 4\pi\hat{\psi}^{(2)}$ describe the linear piezoelectric and piezomagnetic effects. In the limits of the hydrodynamic approach the relations (70) and (71) may be obtained from the thermodynamic potentials as

$$f(E, H, \hat{\sigma}) = -\tfrac{1}{2} E \cdot \hat{\chi} E - \tfrac{1}{2} H \cdot \hat{\kappa} H - \tfrac{1}{2} \hat{\sigma} \cdot \hat{s} \hat{\sigma} - E \cdot \hat{\psi} H - E \cdot \hat{\psi}^{(1)} \hat{\sigma} - H \cdot \hat{\psi}^{(2)} \hat{\sigma} \tag{72}$$

or in the variables p, m, $\hat{u}$ as

$$\begin{aligned} w(p, m, \hat{u}) = {} & \tfrac{1}{2} p \cdot \hat{\chi}^{\circ -1} p + \tfrac{1}{2} m \cdot \hat{\kappa}^{\circ -1} m + \tfrac{1}{2} \hat{u} \cdot \hat{s}^{\circ -1} \hat{u} - p \cdot \hat{\lambda} m - p \cdot \hat{\lambda}^{(1)} \hat{u} \\ & - m \cdot \hat{\lambda}^{(2)} \hat{u}. \end{aligned} \tag{73}$$

E, H and $\hat{\sigma}$ in (72) are independent parameters, the values of which are obtained from the dynamic equations.

The stability of the ground state is defined by a positive definiteness of the quadratic form (73). On approaching the point of phase transition from the side of the symmetric phase its determinant Δ tends to zero and some definite components of the tensors $\hat{\chi}$, $\hat{\kappa}$, $\hat{\psi}^{(1)}$, $\hat{\psi}^{(2)}$ and $\hat{\psi}$ tend to infinity.

Further description is built up the same way as in the previous section. We study in detail some of the most typical examples and then we show the whole set of dipole-active ferroelastic transitions to reduce to one of these cases. As we have already noted, all the qualitatively different situations are realised at the ferroelastic phase transitions along the two-dimensional irreducible representations of the magnetic groups (15) and (16). Therefore, in §§ 3 and 4 we shall repeat the analysis of the dynamics of the softening hydrodynamic modes for the same cases as in the previous section. However, this time the elastic degrees of freedom will also be taken into account sequentially, which will allow us to study the bound electromagnetic and sound waves.

Looking ahead let us note two important circumstances. It turns out that in all the cases allowance for the interaction with the elastic oscillations does not result in any qualitative change of the propagation behaviour of the electromagnetic waves—the whole effect comes from a negligible renormalisation of the velocity of the electromagnetic wave. As for the retroactive effect of the electromagnetic oscillation on the acoustic ones†, it appears that the qualitative changes in the character of sound propagation are observed only in piezo-crystals also having linear magnetoelectric effect. This will be discussed later. Now we shall briefly discuss a situation that happens at dipole-active ferroelastic transitions in crystals without linear magnetoelectric effect. We take as an example the ferroelectric–ferroelastic phase transition, which proceeds along the two-dimensional representation of the point group $D_{2d}\otimes R$ from (15).

When the electromagnetic and transverse sound waves propagate with $q\|z$, only x and y components of the fields, polarisations and displacements arise in a crystal with symmetry $D_{2d}\otimes R$. The corresponding contributions to the thermodynamic potentials (72) and (73) have the form (see table 1)

$$f(E,H,\hat{\sigma}) = -\tfrac{1}{2}\chi(E_x^2+E_y^2) - \tfrac{1}{2}s(\sigma_{xz}^2+\sigma_{yz}^2) - \psi_1(E_x\sigma_{xz}-E_y\sigma_{yz}) - \tfrac{1}{2}\kappa(H_x^2+H_y^2) \tag{74}$$

$$w(p,m,\hat{u}) = \frac{1}{2\chi^\circ}(p_x^2+p_y^2) + \frac{1}{2s^\circ}(u_{x,z}^2+u_{y,z}^2) - \lambda_1(p_xu_{x,z}-p_yu_{y,z}) + \frac{1}{2\kappa^\circ}(m_x^2+m_y^2) \tag{75}$$

where $u_{\alpha,\beta}=\partial u_\alpha/\partial x_\beta$ is the distortion tensor. Besides, $\kappa=\kappa^\circ$; also

$$\chi=\chi^\circ/\xi \qquad s=s^\circ/\xi \qquad \psi_1=\lambda_1\chi^\circ s^\circ/\xi \tag{76}$$

$$\xi = 1-\lambda_1^2\chi^\circ s^\circ = 1-\psi_1^2/\chi s \geqslant 0. \tag{77}$$

On approaching the critical point from the side of the symmetric phase: at $T\to T_c$

$$\xi\to 0 \qquad \chi, s, \psi_1, \varepsilon, \zeta_1 \to \infty. \tag{78}$$

At $q\|z$ the equations (69) for the bound electromagnetic and sound waves split into two independent systems, which describe waves of different polarisation (see the analogous expressions (34) for purely electromagnetic waves)

$$(1)\ E\|p\|x \qquad H\|m\|y \qquad u\|x \tag{79}$$

$$(2)\ E\|p\|y \qquad H\|m\|x \qquad u\|y. \tag{80}$$

Secular equations describing waves of different polarisation have the same forms

$$\left(v^2-\frac{c^2}{\varepsilon\mu}\right)\left(v^2-\frac{1}{\rho s}\right) = \frac{4\pi\psi_1^2 v^4}{\varepsilon s} \tag{81}$$

† In other words we speak here of either electric or magnetic dipole interaction lag effects on the character of sound propagation in crystals with linear piezo-effect.

or in equivalent form

$$\rho s^\circ \frac{\varepsilon^\circ \mu^\circ}{c^2} v^4 - \left(\frac{\mu^\circ}{c^2}(4\pi\chi^\circ + \xi) + \rho s^\circ\right) v^2 + \xi = 0.$$

On approaching T_c from the side of the symmetric phase we have four solutions of the secular equation

$$v_{s\pm} \simeq \pm\left(\frac{\varepsilon\mu}{c^2} + \rho s\right)^{-1/2} = \pm \xi^{1/2}\left(\frac{4\pi\chi^\circ\mu^\circ}{c^2} + \rho s^\circ\right)^{-1/2} \to 0 \tag{82}$$

$$v_{c\pm} \simeq \pm\left(\frac{c^2}{\varepsilon^\circ\mu^\circ} + \frac{4\pi\chi^\circ}{\varepsilon^\circ}\frac{1}{\rho s^\circ}\right)^{1/2} \neq 0. \tag{83}$$

The + and − signs describe waves that propagate in two opposite directions along the z axis. The meaning of the indices s and c will be given below (see formula (85)).

Thus the solution of $v_{s\pm}$ conforms to the softening mode of the bound electromagnetic and sound waves. We have still not used anywhere a supposition on a small value of the sound velocity relative to the velocity of the electromagnetic waves. Let us consider now this rather natural approximation. The given relation is a small parameter

$$\nu = \frac{(\rho s^\circ)^{-1/2}}{c(\varepsilon^\circ\mu^\circ)^{-1/2}} \ll 1. \tag{84}$$

The electrostatic approximation for the sound and the 'frozen lattice' approximation for the electromagnetic wave conform to zero order with respect to ν:

$$v_{s\pm}^{(0)} = \pm\left(\frac{\xi}{\rho s^\circ}\right)^{1/2} = \pm\frac{1}{(\rho s)^{1/2}} \qquad v_{c\pm}^{(0)} = \pm\frac{c}{(\varepsilon^\circ\mu^\circ)^{1/2}}.$$

In the first non-vanishing approximation with respect to ν we get

$$v_{s\pm} \simeq \pm\frac{1}{(\rho s)^{1/2}}\left(1 - \frac{2\pi\psi_1^2\mu}{\rho s^2 c^2}\right) \simeq \pm\left(\frac{\xi}{\rho s^\circ}\right)^{1/2}\left(1 - 2\pi\lambda_1^2\frac{s^\circ\chi^{\circ 2}}{\varepsilon^\circ}\nu^2\right). \tag{85}$$

Thus the difference between the sound velocity in a piezoelectric and the value obtained from the electrostatic approximation does not depend upon the closeness to the critical point and is proportional to an excessively small parameter ν^2. An analogous result was obtained in [13] where, however, the region of phase transition was not studied.

A similar consideration for a piezomagnet leads to the same conclusions. The only difference is confined to the following substitution in the expressions (81)–(85):

$$\mu \leftrightarrow \varepsilon \qquad \chi \leftrightarrow \kappa \qquad \lambda_1 \leftrightarrow \lambda_2 \qquad \psi_1 \leftrightarrow \psi_2. \tag{86}$$

In conclusion to this section let us note that outside the limits of the electrostatic (magnetostatic) approximation when describing the dynamics of the softening mode at the ferroelastic transitions in crystals with linear piezo-effect the situation is not qualitatively changed—as is customary, the velocity of the softening wave decreases at $T \to T_c$ as $\xi^{1/2}$, the only effect being a weak (proportional to ν^2) renormalisation of the velocities of sound and electromagnetic waves. Note that the relative value of this renormalisation does not depend upon the closeness to the critical point. A quite different situation arises at the dipole-active ferroelastic phase transition in a crystal with linear magnetoelectric effect. This problem will be discussed in the next section.

4. Crystals with linear magneto-electric and piezo-effects

This section is central because here we consider the situations when allowance for a lag effect of the electromagnetic interactions qualitatively changes the character of the propagation of the sound wave. These peculiarities are most pronounced near the point of the dipole-active ferroelastic phase transition.

So, we shall continue the interpretation of the peculiarities of the propagation of the bound electromagnetic and sound waves at the phase transitions of the p–m–$\hat{u}$ type. Such transitions are rather common—almost half of the magnetoelectric phase transitions are ferroelastic at the same time. Before we start with particular examples we present simple semi-qualitative considerations based on the results from § 2.

In some cases the presence of a linear magnetoelectric effect may result in the fact that the velocities of the electromagnetic waves in forward and backward directions would not coincide even in the hydrodynamic limits. Assume now that there is a linear binding between the corresponding electromagnetic waves and the sound in such a crystal. The same 'non-reciprocity' could also be expected for the acoustic waves (rigorously speaking only the bound electromagnetic and sound waves may be mentioned here). First, the said effect is electrodynamic in origin, secondly, it does not disappear in the long-wave range $q, \omega \rightarrow 0$ (i.e. it is hydrodynamic) and, finally, it is very convenient for experimental observation. The value of this effect turns out to be proportional to the first power of the small parameter ν from (84).

At ferroelastic and dipole-active phase transitions in crystals without linear magnetoelectric effect the velocity of the softening waves decreases on approaching T_c as $\xi^{1/2}$, where ξ is proportional to the reverse generalised susceptibility appropriate to the order parameter of the phase transition†. Provided this transition is at the same time both dipole-active and ferroelastic we can formally speak of only the softening of the bound electromagnetic and sound waves—the ratio of the sound and electromagnetic waves in the formation of the soft mode does not change at $\xi \rightarrow 0$‡. The situation would change considerably in the presence of linear magnetoelectric effect. According to the results from § 2 the velocity of the softening waves at $T \rightarrow T_c$ may decrease in this case more rapidly—as the first power of ξ. Now we can speak about softening of just electromagnetic waves since the density ρ falls out of the expressions for the velocity of the softening wave at $\xi \rightarrow 0$, which will be described below.

With account taken of the above, let us consider three examples of phase transitions of the p–m–$\hat{u}$ type, which, according to the electrodynamic classification from § 2, refer to the cases 1, 2 and 3. This classification will be preserved here.

4.1. Case 1. Magnetic class $D_{2d}(D_2)$

In the frames of this magnetic group one and the same vector components p and *m* have equal transformation properties (see (19) and table 1), and as a result of p–m–$\hat{u}$ phase transition there appears a polar phase with $p^\circ \| m^\circ$. As it turns out the critical behaviour of the softening branches of the bound electromagnetic and sound waves with $q \| z$ resembles the case for either piezoelectric or piezomagnetic transition in crystals without linear magnetoelectric effect (at $T \rightarrow T_c$, $v_{s\pm}(1) = v_{s\pm}(2) \sim \xi^{1/2}$). This case is the least

† In Landau theory for phase transitions of the second kind $\xi \sim |T - T_c|$.

‡ It is seen in particular from the structure of the expression (83) for the velocity of the softening wave.

interesting and therefore here we shall not give expressions for the corresponding velocities and polarisations.

4.2. *Case 2. Magnetic class* $D_{2d}(C_{2v})$

Instead of (74) and (75) we have the following expressions for the contributions to the thermodynamic potentials $f(E, H, \hat{\sigma})$ and $w(p, m, \hat{u})$, connected with the propagation of the transverse sound and electromagnetic waves with $q \| z$ (see table 1):

$$f(E, H, \hat{\sigma}) = -\tfrac{1}{2}\chi(E_x^2 + E_y^2) - \tfrac{1}{2}\kappa(H_x^2 + H_y^2) - \tfrac{1}{2}s(\sigma_{xz}^2 + \sigma_{yz}^2) - \psi(E_x H_y - E_y H_x) - \psi_1(E_x\sigma_{xz} - E_y\sigma_{yz}) - \psi_2(H_y\sigma_{xz} + H_x\sigma_{yz}) \qquad (87)$$

$$w(p, m, \hat{u}) = \frac{1}{2\chi^\circ}(p_x^2 + p_y^2) + \frac{1}{2\kappa^\circ}(m_x^2 + m_y^2) + \frac{1}{2s^\circ}(u_{x,z}^2 + u_{y,z}^2) - \lambda(p_x m_y - p_y m_x) - \lambda_1(p_x u_{x,z} - p_y u_{y,z}) - \lambda_2(m_y u_{x,z} + m_x u_{y,z}). \qquad (88)$$

The constants present in (87) and (88) are related by the expressions

$$\chi = \frac{\chi^\circ}{\xi}(1 - \lambda_2^2\kappa^\circ s^\circ) \qquad \kappa = \frac{\kappa^\circ}{\xi}(1 - \lambda_1^2\chi^\circ s^\circ) \qquad s = \frac{s^\circ}{\xi}(1 - \lambda^2\chi^\circ\kappa^\circ)$$
$$\psi = \frac{\chi^\circ\kappa^\circ}{\xi}(\lambda + \lambda_1\lambda_2 s^\circ) \qquad \psi_1 = \frac{\chi^\circ s^\circ}{\xi}(\lambda_1 + \lambda\lambda_2\kappa^\circ) \qquad \psi_2 = \frac{\kappa^\circ s^\circ}{\xi}(\lambda_2 + \lambda\lambda_1\chi^\circ) \qquad (89)$$

$$\chi^{\circ-1} = \Delta(\kappa s - \psi_2^2) \qquad \kappa^{\circ-1} = \Delta(\chi s - \psi_1^2) \qquad s^{\circ-1} = \Delta(\kappa\chi - \psi^2)$$
$$\lambda = \Delta(\psi s - \psi_1\psi_2) \qquad \lambda_1 = \Delta(\psi_1\kappa - \psi\psi_2) \qquad \lambda_2 = \Delta(\psi_2\chi - \psi\psi_1) \qquad (90)$$

where

$$\xi = 1 - \lambda^2\kappa^\circ\chi^\circ - \lambda_1^2\chi^\circ s^\circ - \lambda_2^2\kappa^\circ s^\circ - 2\lambda\lambda_1\lambda_2\chi^\circ \geqslant 0$$
$$\Delta = \frac{\xi}{\chi^\circ\kappa^\circ s^\circ} = (\chi\kappa s - \psi^2 s - \psi_1^2\kappa - \psi_2^2\chi + 2\psi\psi_1\psi_2)^{-1}. \qquad (91)$$

At $T \to T_c$, $\xi \to 0$ and all the parameters from (87) also diverge according to (89).

If we put $\lambda = \lambda_2 = 0$, then the expressions (87)–(91) coincide with the corresponding formulae from the previous section, dedicated to piezoelectrics (or to piezomagnetics at $\lambda = \lambda_1 = 0$). If we put $\lambda_1 = \lambda_2 = 0$ the whole consideration will be reduced to case 2 from § 2.

The dynamic equations (69) for bound electromagnetic and sound waves with account of relations resulting from (87) also split at $q \| z$ into two separate systems, which describe the waves with different polarisations (see expressions (79) and (80)).

The secular equations corresponding to both polarisations have the same form

$$\left(v^2 - \frac{(c - \zeta v)^2}{\varepsilon\mu}\right)(v^2 - 1/\rho s) = 8\pi\frac{\psi_1\psi_2}{\varepsilon\mu s}(c - \zeta v)v^3 + 4\pi\left(\frac{\psi_1^2}{\varepsilon s} + \frac{\psi_2^2}{\mu s}\right)v^4. \qquad (92)$$

Equation (92) has four different roots describing for each polarisation (79) and (80) two bound electromagnetic and sound waves propagating in two opposite directions along the z axis.

In consequence of § 2 the forward and backward directions cannot be equivalent;

that is, for each polarisation 1 or 2

$$\begin{aligned} v_{s+}(1) &= v_{s+}(2) \neq -v_{s-}(1) = -v_{s-}(2) \\ v_{c+}(1) &= v_{c+}(2) \neq -v_{c-}(1) = -v_{c-}(2) \end{aligned} \tag{93}$$

where as before + and − denote two opposite directions of wave propagation with $q \| z$. Indices s and c have a simple physical sense in the ultimate case, where it is possible to consider separately in a good approximation the sound (s) and the light (c).

When approaching T_c from the side of the symmetry phase one of four roots from equations (92) turns to zero: at $T \rightarrow T_c$

$$\frac{v}{c} \simeq \frac{\xi}{8\pi\chi^\circ\kappa^\circ(\lambda + \lambda_1\lambda_2 s^\circ)} \simeq \frac{1}{2\zeta} \simeq \frac{\operatorname{sgn}\zeta}{2(\varepsilon\mu)^{1/2}}. \tag{94}$$

Thus, in case 2 the velocity of the softening mode at $T \rightarrow T_c$ decreases much more rapidly than in the case discussed in the previous section. Note that, since the density ρ vanishes from the asymptotic expression (94) for the velocity, one can state that in this case the role of the softening mode formally belongs to the electromagnetic waves. As for the elastic deformations, in the limits of (94) they succeed in a quasi-static manner to adapt to the fields and polarisations existing in an electromagnetic wave. Therefore, it is natural that the expression (94) is in agreement with the corresponding expression (39), which is related to the case of a purely electromagnetic wave. In particular, the direction of propagation of the soft mode (the direction + or − along the z axis) is defined by the sign of ζ. Note one more essential circumstance, which is not closely associated with a phase transition. We shall discuss the violation of the equality of the velocities of transverse sound propagating in two opposite directions along the z axis. In the first non-vanishing approximation with respect to a small parameter ν from (84) we obtain from (92) for this difference

$$\Delta v_s = v_{s+} + v_{s-} \simeq \frac{8\pi\psi_1\psi_2}{\rho s^2 c} = \frac{8\pi}{\rho c}\chi^\circ\kappa^\circ \frac{(\lambda_1 + \lambda\lambda_2\kappa^\circ)(\lambda_2 + \lambda\lambda_1\chi^\circ)}{(1 - \lambda^2\kappa^\circ\chi^\circ)^2} \tag{95}$$

where in the zero approximation with respect to ν

$$v_{s+}^{(0)} = -v_{s-}^{(0)} = (\rho s)^{-1/2}.$$

The effect in (95), unlike the electrodynamic correction in (85), is linear with respect to ν and consequently much stronger. Moreover, in this case not only a correction is meant but also a qualitative change in the character of the sound propagation—the violation of the reciprocity principle in the formulation, presented in § 2. Such an effect can easily be observed in an experiment.

4.3. Case 3. Magnetic class D_{2d}

Here instead of (87) and (88) we have the following expressions for the thermodynamic potentials f and w related to the propagation of sound and electromagnetic waves

$$\begin{aligned} f(E, H, \hat\sigma) = &-\tfrac{1}{2}\chi(E_x^2 + E_y^2) - \tfrac{1}{2}\kappa(H_x^2 + H_y^2) - \tfrac{1}{2}s(\sigma_{xz}^2 + \sigma_{yz}^2) - \psi(E_xH_y + E_yH_x) \\ &- \psi_1(E_x\sigma_{xz} - E_y\sigma_{yz}) - \psi_2(H_y\sigma_{xz} - H_x\sigma_{yz}). \end{aligned}$$

$$\begin{aligned} w(p, m, \hat u) = &\frac{1}{2\chi^\circ}(p_x^2 + p_y^2) + \frac{1}{2\kappa^\circ}(m_x^2 + m_y^2) + \frac{1}{2s^\circ}(u_{x,z}^2 + u_{y,z}^2) - \lambda(p_xm_y + p_ym_x) \\ &- \lambda_1(p_xu_{x,z} - p_yu_{y,z}) - \lambda_2(m_yu_{x,z} - m_xu_{y,z}). \end{aligned}$$

Relations (89)–(91) hold here.

Here, as in case 2, the dynamic equations with account taken of substantial relations at $q \| z$ split into non-bound sets of equations, which describe the waves with polarisations 1 from (79) and 2 from (80). Note that the equations describing the waves of the first polarisation are exactly equivalent to the corresponding equations of the magnetic class $D_{2d}(C_{2v})$ (case 2), whereas the equations describing the waves of the second polarisation differ from those of case 2 only in the substitution

$$\psi \to -\psi \qquad \psi_2 \to -\psi_2. \tag{96}$$

According to (89)–(91) there should also be the substitution

$$\lambda \to -\lambda \qquad \zeta \to -\zeta \qquad \lambda_2 \to -\lambda_2 \qquad \zeta_2 \to -\zeta_2.$$

Thus, the secular equation describing the waves of the first polarisation coincide with (92) and for the waves with the second polarisation it differs from (92) by the substitution (96). It is easily seen that substitution (96) in equation (92) is equivalent to the substitution $v \to -v$. Therefore, in the considered case 3 we have instead of (93) for every polarisation 1 or 2

$$\begin{aligned} v_{s+}(1) &= -v_{s-}(2) \neq v_{s+}(2) = -v_{s-}(1) \\ v_{c+}(1) &= -v_{c-}(2) \neq v_{c+}(2) = -v_{c-}(1). \end{aligned} \tag{97}$$

The expressions (94) and (95) as well as all the comments hold here (with account of the substitution (96) for the waves of the second polarisation).

The formulae (97) are analogous to the relations (65). The reciprocity principle for the sound in the wide sense is satisfied here; however for each polarisation the wave velocities 'forwards' and 'backwards' differ in magnitude, and therefore in the narrow sense the reciprocity principle is violated.

4.4. Case 4. Magnetic class $D_{2d}(S_4)$

This case differs from the previous one only in the fact that instead of (79) and (80) the normal modes with $q \| z$ have the following polarisations

$$\begin{aligned} &(1)\ E_x = E_y && H_x = -H_y && \sigma_{xz} = -\sigma_{yz} \\ &(2)\ E_x = -E_y && H_x = H_y && \sigma_{xz} = \sigma_{yz} \end{aligned}$$

(and similarly for the components p, m and $\dot{u}$). The relations (97) and all comments hold true here. This is the reason for the magnetic classes $D_{2d}(S_4)$ and D_{2d} to be related to case 3 according to the classification in tables 1 and 2. As before, the velocity of the soft mode near the critical point is defined by a purely electrodynamic expression (94). There are two waves that soften: the first with polarisation 1, which propagates in one of the directions along the z axis, and the second with polarisation 2, propagating in the opposite direction.

Let us note now one significant difference between the magnetic classes D_{2d} and $D_{2d}(S_4)$ concerning their acoustic properties. The matter is that the above consideration concerns only either non-polarised crystals (i.e. crystals for which the equilibrium values of the electric p° and magnetic m° polarisations are zero) or at least cases where the intensity of the homogeneous part of the electric and magnetic fields (E° and H° respectively) is zero in the crystal. Otherwise, allowance for the effects resulting from violated rotational invariance of the energy density of the crystal becomes necessary. The corresponding effects are rather thoroughly analysed in [1], so we do not return to the problem. Note only that, in the case of the polarised crystals at E° or H° being non-zero, the velocity of the softening waves in a critical point depends upon mechanical boundary conditions, since the boundary conditions influence the position of the critical point itself. Finally, in magnetically polarised crystals (case 4) at rather strong external field, the Lorentz force $(1/c)[J^{\circ} \times B] \rightarrow (1/c)[p^{\circ} \times B]$ should be taken into account in the right part of the equation (68) for motion.

4.5. Summary

A brief outline of the main results of the fourth section is given below.

In crystals with linear piezo-effect the number of softening branches of the spectrum of the bound acoustic and electromagnetic waves is less than it could be without allowance for the connection of polarisation with deformation. In particular, for transitions of the *p–m–û* type in the last column of table 2 the wavevectors q^*, to which the softening modes conform, are given. From the comparison of the fourth and last columns it is seen that allowance for the piezo-effect decreases the number of directions to which the softening modes for all the cases in table 2(*c*) conform. In some cases from table 2(*a*) the softening modes completely disappear†.

If without allowance for the piezo-effect the velocity of the softening electromagnetic waves tends to zero linearly along ξ (which is possible only for the *p–m–û* transitions from table 2), first, its account will preserve this dependence and, secondly, at rather small ξ we can speak of the fact that it is the electromagnetic waves that soften but not the sound (the density ρ is absent in the expressions for the velocity of the softening branch). But if the velocity of the electromagnetic waves without allowance for the piezo-effect decreases at $T \rightarrow T_c$ as $\xi^{1/2}$, we can formally speak only about the softening of the bound elastic and electromagnetic waves.

5. Conclusions

Summing up our considerations of the dipole-active and ferroelastic phase transitions in dielectrics we shall emphasise once more that the behaviour of the softening electromagnetic or sound waves differs in the main from the 'classical' one (see formulae (1) and the corresponding comments) only in crystals with linear magnetoelectric effect. This circumstance is reflected in the title of the paper. The corresponding anomalies are described rather in detail in §§ 2 and 4. As for conducting crystals, a quite different picture appears here. We present a brief summary of the problem.

In the case of plane transverse waves, which conform to the condition (3), allowance for electrical conduction reduces to the following substitution in the Maxwell equations (26):

$$\hat{\varepsilon} \rightarrow \hat{\varepsilon} + \mathrm{i}4\pi\hat{\sigma}/\omega$$

where $\hat{\sigma}$ is the tensor of the electrical conductivity. An analogous substitution should be made in all the dynamic equations considered before, describing the softening electromagnetic as well as bound electromagnetic and sound waves. Analysis of the corresponding dispersion equations falls outside the limits of the present paper. An independent paper will be dedicated to this problem [14]. Now we restrict ourselves to quoting some basic conclusions.

In the low-frequency limits $4\pi\sigma/\varepsilon\omega \gg 1$, the electromagnetic waves cannot propa-

† See footnote at very end of § 2.

gate in the substance† (see for example [8]). Therefore we shall confine ourselves to the consideration of acoustic waves in crystals with linear piezo-effect.

In conducting piezoelectrics the character of the propagation of transverse sound does not differ practically from the case of dielectrics. But in the region of the ferroelastic (piezoelectric) phase transition even such a negligible difference vanishes.

In a conducting piezomagnet the velocity of transverse sound in the low-frequency limits is defined by the expression

$$v_s^2 \simeq (\rho s)^{-1} \frac{\mu}{\mu^\circ} \qquad \text{at} \qquad \frac{4\pi\sigma}{\omega} \gg \frac{c^2/\varepsilon\mu}{1/\rho s}.$$

On approaching the point of the piezomagnetic phase transition:

$$\mu \sim s \sim \xi^{-1} \to \infty.$$

With this, the velocity of the low-frequency sound v_s remains finite: at $T \to T_c$

$$v_s^2 \to \frac{4\pi\kappa^\circ}{\rho s^\circ \mu^\circ} > 0.$$

The role of the 'softening' mode passes to a fully damped electromagnetic wave with the quadratic law of dispersion: at $T \to T_c$

$$\omega \simeq -\mathrm{i}q^2 \frac{c^2}{4\pi\sigma\mu} \simeq -\mathrm{i}q^2\xi \frac{c^2}{(4\pi)^2\sigma\kappa^\circ}.$$

A similar situation also occurs in conducting piezomagnetoelectrics.

Naturally, all these conclusions refer to purely transverse waves—just they are softening in all cases of our interest. But if the condition (3) is not satisfied, then significantly stronger electrostatic effects, connected with the advent of bulk charge density in the sound wave, will advance forward in piezoelectrics and piezomagnetoelectrics [8, 13].

References

[1] Bar'yakhtar V G, Vitebskii I M, Lavrinenko N M and Sobolev V L 1986 *Zh. Eksp. Teor. Fiz.* **90** 1111–17
[2] Bar'yakhtar V G, Vitebskii I M, Lavrinenko N M and Sobolev V L 1988 *Preprint* ITP-112 R, Institute for Theoretical Physics, Ukrainian Academy of Sciences, Kiev
[3] Krivoglaz M A 1963 *Fiz. Tverd. Tela* **5** 3439–52
[4] Vitebskii I M, Zeltzer A S and Lavrinenko N M 1988 *Ukr. Fiz. Zh.* **33** 892–7
[5] Cowley R A 1976 *Phys. Rev.* B **13** 4877–85
[6] O'Dell T 1970 *The Electrodynamics of Magnetoelectric Media* (Amsterdam: North-Holland)
[7] Freeman A and Schmid H (ed.) 1973 *Magnetoelectric Interaction Phenomena in Crystals* (New York: Gordon and Breach)
[8] Landau L D and Lifshitz E M 1982 *Electrodynamics of Continuous Media* (Moscow: Nauka) p 624
[9] Sirotin Yu I and Shaskol'skaya M P 1979 *Fundamental Crystallophysics* (Moscow: Nauka) p 639
[10] Fush R 1965 *Phil. Mag.* **11** 647–9
[11] Khmel'nitskii D E 1974 *Fiz. Tverd. Tela* **16** 3188–90
[12] Courtens E and Gammon R 1981 *Phys. Rev.* B **24** 3890–902
[13] Hudson A R and White D L 1962 *J. Appl. Phys.* **33** 40–7
[14] Vitebskii I M, Zeltzer A S, Lavrinenko N M and Sobolev V L 1989 *Fiz. Nizk. Temp.* **15** 750–8

† Excluded are magnetically polarised conductors, in which the tensor of the electrical conductivity has the antisymmetric (Hall) part. This case is not considered here.