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1989 J. Phys.: Condens. Matter 1 79

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Ferroelastic phase transitions in polarised media

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Received 4 February 1988, in final form 13 May 1988

Abstract. The critical behaviour under ferroelastic phase transitions in polarised crystals is studied. The fundamental role of the effects due to the violation of the rotational invariance of the energy of a system in a magnetic or electric field as well as of the effects connected with the non-local character of dipole interactions is elucidated. The acoustic properties of polarised crystals in the limit of small wavevectors are investigated using the magnetostatic (or electrostatic) approach.

1. Introduction

Ferroelastic phase transitions constitute rather a wide range of magnetic and structural phase transitions in solids. From the symmetry point of view the proper ferroelastic transition is a transition whose order parameter is linearly connected with non-isomorphic striction, i.e. with macroscopic deformations of the crystal lattice. This connection is known to cause anomalies of elastic properties of these systems and to determine, to a great extent, peculiarities of their critical behaviour in the most general sense (Patashinski and Pokrovskii 1982, Cowley 1976, Schwabl 1985, David 1983, Villain 1970, Levanuk and Sobianin 1970, Fossum 1985). This paper is devoted to the investigation of these peculiarities for the case of ferroelastic phase transitions in polarised crystals, i.e. in crystals possessing either a magnetic or electric dipole moment. Polarisation can be either spontaneous (pyroelectrics and ferromagnets) or induced by external field. Peculiarities of the critical behaviour which will be considered below have a completely general character and do not depend on particular physical causes of the phase transition, i.e. they do not depend on the microscopic realisation of the order parameter. The main cause of these peculiarities in polarised media is the violation of invariance of the crystal energy with respect to its orientation in the presence of an external field. The present paper is intended to analyse these problems within a purely thermodynamic approach without application to any model assumption (Bar'yakhtar *et al* 1986a, b).

The acoustic vibration spectrum of an infinite crystal is activationless; therefore at sufficiently small wavevectors q , vibrations in a sound wave are so slow that any subsystem (e.g. a magnetic one) has time to adjust itself to the field of deformation arising in the sound wave. This is associated with the fact that at $q \rightarrow 0$ the velocity of sound is determined by the static elastic moduli of the crystal which are formed with the

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participation of all subsystems interacting with the deformations (the hydrodynamic principle of local equilibrium). It is this region of the acoustic vibration spectrum which we shall study below. At large wavevectors the approach based on continuum elasticity theory is insufficient, because it becomes necessary to write explicit dynamic equations for various internal degrees of freedom which interact with the sound; this goes beyond the local equilibrium principle and requires application of model assumptions (Melcher 1972, Akhiezer *et al* 1967, Vlasov 1962, Chow and Keffer 1973, Brown 1965, Bar'yakhtar and Turov 1985, Bar'yakhtar *et al* 1986a, b, Maugin and Eringen 1972, Zhelnorovich 1987, Vendik and Mironenko 1974, Sedov 1970, de Groot and Mazur 1962).

In the absence of magnetic and electric fields the energy density of a weakly deformed crystal is determined by the symmetric deformation tensor (Landau and Lifshitz 1965):

$$w = \frac{1}{2} c_{\alpha\beta, \mu\nu} u_{\alpha\beta} u_{\mu\nu}. \quad (1)$$

Here and below components $u_{\alpha\beta}$ of the deformation tensor describe small deviations from the equilibrium position arising in a sound wave. The same is valid for other components of the distortion tensor:

$$u_{\alpha, \beta} = \partial u_{\alpha} / \partial x_{\beta} = u_{\alpha\beta} + \omega_{\alpha\beta} \quad (2)$$

where

$$u_{\alpha\beta} = \frac{1}{2}(u_{\alpha, \beta} + u_{\beta, \alpha}) \quad \omega_{\alpha\beta} = \frac{1}{2}(u_{\alpha, \beta} - u_{\beta, \alpha}).$$

To derive equation (1), we used two assumptions.

(i) w is invariant with respect to the orientation of the body volume element in a space (rotational invariance).

(ii) Long-range (Coulomb and dipole) interactions are absent.

In the presence of either a magnetic or an electric field, both these assumptions are violated.

At the point of ferroelastic transition the proper quadratic form (1) loses positive definiteness, and anomalies due to the critical behaviour are completely determined by anomalies of the crystal elastic properties. Here we represent rather a simple reasoning to explain this conclusion.

In the absence of external fields the expansion for the energy in powers of order parameter η and deformation tensor $\hat{\mathbf{u}}$ at the ferroelastic transition has the following structure:

$$w(\eta, \hat{\mathbf{u}}) = w^{(0)}(\eta) + w^{(0)}(\hat{\mathbf{u}}) - \lambda_{i, \mu\nu} \eta_i u_{\mu\nu} \quad (3)$$

where

$$w^{(0)}(\eta) = \frac{1}{2} a^{(0)} \eta_i \eta_i + \dots \quad w^{(0)}(\hat{\mathbf{u}}) = \frac{1}{2} c_{\alpha\beta, \mu\nu}^{(0)} u_{\alpha\beta} u_{\mu\nu}. \quad (4)$$

Here η_i are the order parameter components (the index i enumerates the lines of irreducible representation responsible for the phase transition); $c_{\alpha\beta, \mu\nu}^{(0)}$ are the trial elastic moduli (the structures of the expressions for $w^{(0)}(\hat{\mathbf{u}})$ and $w(\hat{\mathbf{u}})$ from (1) are exactly the same).

Now let us change from the Cartesian components $u_{\alpha\beta}$ of the deformation tensor to their linear combination fulfilling irreducible representations of the symmetry group of high-symmetry phase

$$u_{\mu\nu} = \xi_{i, \mu\nu}^{(\Gamma)} u_i^{(\Gamma)}.$$

Here Γ is the number of irreducible representations and i is the index of its line. Rewriting (3) and (4) in the new variables, we obtain

$$w(\eta, \hat{\mathbf{u}}) = w^{(0)}(\eta) + w^{(0)}(\hat{\mathbf{u}}) - \lambda \eta_i u_i \quad (5)$$

where

$$w^{(0)}(\hat{\mathbf{u}}) = \frac{1}{2} c^{(0)} u_i u_i + \dots \quad (6)$$

The trial elastic energy (6) presents in a detailed form only the term corresponding to irreducible representation by which the order parameter ($u_i \sim \eta_i$) is transformed. The index Γ for this representation is omitted. The value of $c^{(0)}$ in (6) is the corresponding linear combination of the elastic moduli $c_{\alpha\beta, \mu\nu}^{(0)}$.

Using the condition $\partial w(\eta, \hat{\mathbf{u}})/\partial \eta = 0$, we eliminate the variables η_i from equation (5). This yields equation (1) for the free-energy density

$$w(\hat{\mathbf{u}}) = \frac{1}{2} c_{\alpha\beta, \mu\nu} u_{\alpha\beta} u_{\mu\nu} \equiv \frac{1}{2} c u_i u_i + \dots \quad (7)$$

where

$$c = c^{(0)} - \lambda^2/a^{(0)}.$$

If, by using the condition $\partial w(\eta, \hat{\mathbf{u}})/\partial \hat{\mathbf{u}} = 0$, we eliminate the deformation tensor components from (5), we then obtain

$$w(\eta) = \frac{1}{2} a \eta_i \eta_i + \dots \quad (8)$$

where

$$a = a^{(0)} - \lambda^2/c^{(0)}.$$

The symmetry phase stability region is determined by one of equivalent inequalities

$$a \geq 0 \quad \text{or} \quad c \geq 0. \quad (9)$$

At the ferroelastic transition point

$$a = 0 \quad (c = 0) \quad (10)$$

the quadratic forms (1), (3), (5), (7) and (8) simultaneously lose their positive definiteness.

Deformation arising in a sound wave at $\mathbf{q} \rightarrow \mathbf{0}$ is adiabatic; therefore, throughout the paper, only adiabatic elastic moduli are used. However, since the elastic modulus c from (7) and (10) determines the energy of the non-isomorphic irreducible deformation (corresponding to purely transverse sound), the adiabatic and isothermal values of this modulus in the symmetric phase coincide.

Critical peculiarities of the elastic properties (in particular, the critical behaviour of a long-wavelength sound) can be analysed using the equivalent expressions (1) or (7) for the true (renormalised) elastic energy. We recall once again that these expressions are valid, strictly speaking, only in the absence of elastic and magnetic fields since, in their derivation, the use was made of invariance of the energy density with respect to the orientation of the body volume element in a space—even in the presence of homogeneous fields, this invariance either partially or completely (for crossed electric and magnetic fields) disappears. Prior to the start of the discussion on the effects associated with the violated rotational invariance, it should be stated which peculiarities in the sound behaviour appear at proper ferroelastic transitions in non-polarised crystals without piezo-electric and piezomagnetic effects.

The combined analysis of the stability (positive definiteness) of the quadratic form (1) or (7) and expressions for the velocities of long-wavelength sound in crystals of various syngonies show the following in all cases.

(i) At the critical point (10) the velocity of either one or two branches of transverse sound propagating in certain directions becomes zero (Cowley 1976).

Then the direct consequence of (i) is the so-called reciprocity principle consisting of the equality of the velocities of the transverse sound with the x polarisation propagating along the y axis and that with y polarisation propagating along the x axis (the x and y directions are perpendicular to each other). Thus, in addition to (i), the following statement is true.

(ii) If the velocity of the x -polarised transverse sound wave with $q \parallel y$ has become zero, the same will happen to the y -polarised wave with $q \parallel x$.

In the above case the peculiarities of the critical properties are unambiguously determined by the symmetry of the initial phase and by the transformation properties of the order parameter of the proper ferroelastic transition. In fact, the macroscopic symmetry of the initial phase and the number of irreducible representations responsible for the ferroelastic phase transition unambiguously determine the peculiarities of elastic properties (including long-wavelength sound) in the critical region. It is elastic subsystem that is abnormally fluctuating in this case. A similar situation is typical of the phase transition theory.

However, for proper ferroelastic phase transitions in polarised media (in the presence of either magnetic or electric fields), this statement is already invalid. We shall see below that, for the second-order phase transition in a polarised medium, the critical behaviour in various cases can be different even when there is the same symmetry of the order parameter. This is connected with the violation of rotational invariance of the energy density ω in the presence of the field.

As was mentioned above, acoustic vibrations in the small-wavevector q limit can be considered within the continuum elasticity theory, the velocity of sound being determined by the same parameters (adiabatic moduli) as the static elastic properties. However, equation (1) for the energy density in the presence of the field is not acceptable since it was derived by assuming rotational invariance, and violation of this invariance results in the appearance of anti-symmetric part $\omega_{\alpha\beta}$ of the distortion tensor in the expression for w . Dependence of density energy on ω qualitatively changes the character of critical behaviour of ferroelastic phase transition. Furthermore the character of the sound propagation is greatly influenced by the long-range dipole interaction which is always present in polarised media as well as in crystals possessing linear piezo-electric or piezomagnetic properties (Landau and Lifshitz 1982). All these peculiarities will be considered in the following two sections within the consistent approach framework. For definiteness, we shall deal with magnetic polarisation and magnetic field. The cases of electric polarisation and electric field differ from the above one only by differences in notation (naturally, if a dielectric medium is meant). Everywhere in this paper the magnetostatic approximation is used. Strictly speaking, for a closed description of acoustic vibrations in polarised media and media with a linear piezomagnetic effect, even at $q \rightarrow 0$ one should supplement the elasticity theory equations with equations of continuum electrodynamics. However, taking into account the effects of electromagnetic interaction lagging does not change the results obtained below considerably. We shall return to this question at the end of this paper.

2. Effective elastic energy of a sound wave in a polarised medium

In view of the above considerations, we proceed from the following expression for the potential energy of a magnetically polarised medium (see, e.g., Akhiezer *et al* 1967):

$$W = \int d^3x (w_i - \mathbf{M} \cdot \mathbf{H}_{\text{ext}} - \frac{1}{2} \mathbf{M} \cdot \mathbf{H}_m) \quad (11)$$

where w_i is the density of internal energy which is a function of the magnetisation \mathbf{M} and distortion tensor $u_{\alpha, \beta}$. The second and third terms in the integrand describe the Zeeman energy (\mathbf{H}_{ext} is the external field) and the dipolar interaction energy (\mathbf{H}_m is the dipolar field), respectively.

The internal energy density w_i does not depend on the orientation of the crystal volume element in a space. The Zeeman term $-\mathbf{M} \cdot \mathbf{H}_{\text{ext}}$ in (11) is responsible for violation of the rotational invariance of total energy W . The dipolar contribution to W possesses only global rotational invariance since the dipolar field \mathbf{H}_m at the fixed point depends on the magnetisation distribution throughout the whole crystal. The qualitative peculiarities of long-wavelength sound propagation in polarised media are determined by the absence of local rotational invariance of the latter two contributions to the energy in (11).

Let us determine the contribution to the energy (11) associated with the propagation of a small-amplitude sound wave. For this purpose, we expand each term of the integrand in a power series of small deviations from the equilibrium position, i.e. in a series of

$$\mathbf{m} = \mathbf{M} - \dot{\mathbf{M}}; u_{\alpha, \beta} \quad (12)$$

where $\dot{\mathbf{M}}$ is the equilibrium polarisation. Taking into consideration the rotational invariance of the function w_i , we have, in a harmonic approximation,

$$w_i = \dot{w} + \boldsymbol{\mu} \cdot \dot{\mathbf{H}} + \frac{1}{2} \boldsymbol{\mu} \cdot \hat{\boldsymbol{\chi}}^{-1} \boldsymbol{\mu} - \boldsymbol{\mu} \cdot \hat{\boldsymbol{\lambda}} \dot{\mathbf{u}} + w^{(0)}(\dot{\mathbf{u}}) \quad (13)$$

where

$$\boldsymbol{\mu} = \hat{\mathbf{R}}^{-1} \mathbf{M} - \dot{\mathbf{M}} = \hat{\mathbf{R}}^{-1} \mathbf{m} + (\hat{\mathbf{R}}^{-1} - \hat{\mathbf{I}}) \dot{\mathbf{M}}. \quad (14)$$

The rotation operator $\hat{\mathbf{R}}$ in the Euler representation is expressed in terms of the distortion tensor component (Melcher 1972) as follows:

$$\begin{aligned} \hat{\mathbf{R}} &= [\hat{\mathbf{I}} - 2\dot{\mathbf{u}} + (\dot{\mathbf{u}} - \dot{\boldsymbol{\omega}})(\dot{\mathbf{u}} + \dot{\boldsymbol{\omega}})]^{1/2} (\hat{\mathbf{I}} - \dot{\mathbf{u}} - \dot{\boldsymbol{\omega}})^{-1} \\ &= \hat{\mathbf{I}} + \dot{\boldsymbol{\omega}} + \frac{1}{2} \dot{\boldsymbol{\omega}}^2 + \frac{1}{2} (\dot{\mathbf{u}} \dot{\boldsymbol{\omega}} + \dot{\boldsymbol{\omega}} \dot{\mathbf{u}}) + O(u_{\alpha, \beta}^3). \end{aligned} \quad (15)$$

The geometrical meaning of the value of $\boldsymbol{\mu}$ is simple, namely it is the deviation of polarisation from the equilibrium value in the reference system locally connected with the crystallographic axes. According to (14) and (15),

$$\boldsymbol{\mu} = \mathbf{m} - \dot{\boldsymbol{\omega}} \dot{\mathbf{M}} + \frac{1}{2} (\dot{\boldsymbol{\omega}}^2 - \dot{\mathbf{u}} \dot{\boldsymbol{\omega}} - \dot{\boldsymbol{\omega}} \dot{\mathbf{u}}) \dot{\mathbf{M}} - \dot{\boldsymbol{\omega}} \mathbf{m} + \dots$$

Replacement of \mathbf{m} in (13) by $\boldsymbol{\mu}$ (i.e. by a value which does not change on rotation of the crystal volume element) automatically provides rotational invariance of the corresponding terms of the expansion of w_i . The meaning of the other values of (13) is as follows. $\hat{\boldsymbol{\chi}}$ is the magnetic susceptibility tensor at $u_{\alpha, \beta} = 0$. The third-rank tensor $\hat{\boldsymbol{\lambda}}$ is connected with the piezomodulus tensor $\hat{\mathbf{d}}$ by the relationship†

$$\hat{\mathbf{d}} = \hat{\boldsymbol{\chi}} \hat{\boldsymbol{\lambda}} \quad (\hat{d}_{\alpha, \beta \gamma} = \chi_{\alpha \mu} \lambda_{\mu, \beta \gamma}). \quad (16)$$

† In accordance with the comments made after equation (10), the material tensors $\hat{\boldsymbol{\chi}}$, $\hat{\boldsymbol{\lambda}}$ and $\hat{\mathbf{d}}$ are adiabatic.

$w^{(0)}(\hat{\mathbf{u}})$ is the trial elastic energy equal, by definition, to $w_i - \dot{w}$ at $\boldsymbol{\mu} = \tilde{\mathbf{0}}$ (in the harmonic approximation, $w^{(0)}$ depends only on the symmetric part of the distortion tensor $u_{\alpha,\beta}$ since each term in equation (13) (it concerns $w^{(0)}$ too) should have rotational invariance). Finally, $\hat{\mathbf{H}}$ coincides with the strength of the internal magnetic field \mathbf{H} . For an ellipsoidal specimen,

$$\mathbf{H} = \mathbf{H}_{\text{ext}} - 4\pi\hat{\mathbf{N}}\dot{\mathbf{M}} \quad (17)$$

where $\hat{\mathbf{N}}$ is the tensor of demagnetising coefficients. Naturally, the parameters $\dot{\mathbf{M}}$, $\hat{\boldsymbol{\chi}}$ and $\hat{\boldsymbol{\lambda}}$ as well as the 'trial moduli' $c^{(0)}$ in the expression for $w^{(0)}$ and, finally, the uniform equilibrium deformation (which is used as a reference point for the non-uniform deformations $u_{\alpha,\beta}$ appearing in the sound wave) and, consequently, the tensor components are functions of \mathbf{H} . In particular, at $\mathbf{H} = \mathbf{0}$ the values $\dot{\mathbf{M}}$ and $\hat{\boldsymbol{\lambda}}$ can also become zero for either a paramagnet or an antiferromagnet without the linear piezomagnetic effect.

Dipolar interaction (the last term in (11)), firstly, leads to a demagnetising field which is reduced to the replacement of \mathbf{H}_{ext} by \mathbf{H} from (17) in the Zeeman energy (Akhiezer *et al* 1967). Secondly, additional energy w_m appears, which is connected with non-homogeneous part \mathbf{h} of the dipolar field \mathbf{H}_m . For a plane wave, we have

$$\mathbf{h} = -4\pi n(\mathbf{n} \cdot \mathbf{m}) \quad w_m = 2\pi(\mathbf{n} \cdot \mathbf{m})^2 \quad (18)$$

where $\mathbf{n} = \mathbf{q}/|\mathbf{q}|$ is the unit vector coinciding with the plane-wave propagation direction.

From equations (13)–(15), (17) and (18), we obtain the following expression for the plane-wave energy density (neglecting the kinetic energy $\rho\dot{u}^2/2$ where ρ is the material density):

$$\begin{aligned} w(\mathbf{m}, \hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}) = & \dot{w} + \frac{1}{2}(\mathbf{m} - \hat{\boldsymbol{\omega}}\dot{\mathbf{M}}) \cdot \hat{\boldsymbol{\chi}}^{-1}(\mathbf{m} - \hat{\boldsymbol{\omega}}\dot{\mathbf{M}}) - (\mathbf{m} - \hat{\boldsymbol{\omega}}\dot{\mathbf{M}}) \cdot (\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + 2\pi(\mathbf{n} \cdot \mathbf{m})^2 \\ & + w^{(0)}(\hat{\mathbf{u}}) - \frac{1}{2}\dot{\mathbf{M}} \cdot \hat{\boldsymbol{\omega}}^2\mathbf{H} + \dot{\mathbf{M}} \cdot [\hat{\boldsymbol{\omega}} + (\hat{\mathbf{u}}\hat{\boldsymbol{\omega}} + \hat{\boldsymbol{\omega}}\hat{\mathbf{u}})/2]\mathbf{H}. \end{aligned} \quad (19)$$

For the long-wavelength sound value, \mathbf{m} adjusts itself to the instantaneous values $\hat{\mathbf{u}}$ and $\hat{\boldsymbol{\omega}}$ in a quasi-static way. Thus, from the condition of min w with respect to \mathbf{m} , we have

$$\mathbf{m} = \hat{\boldsymbol{\omega}}\dot{\mathbf{M}} + \hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) - [4\pi/K(n)]\hat{\boldsymbol{\chi}}n\{\mathbf{n} \cdot [\hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + \hat{\boldsymbol{\omega}}\dot{\mathbf{M}}]\} \quad (20)$$

where

$$K(n) = 1 + 4\pi n \cdot \hat{\boldsymbol{\chi}}n.$$

Eliminating \mathbf{m} from equation (19), we obtain finally

$$\begin{aligned} w(\hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}) = & -\frac{1}{2}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) \cdot \hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + w^{(0)}(\hat{\mathbf{u}}) - \frac{1}{2}\dot{\mathbf{M}} \cdot \hat{\boldsymbol{\omega}}^2\mathbf{H} + [2\pi/K(n)] \\ & \times \{\mathbf{n} \cdot [\hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + \hat{\boldsymbol{\omega}}\dot{\mathbf{M}}]\}^2 + \dot{\mathbf{M}} \cdot [\hat{\boldsymbol{\omega}} + (\hat{\mathbf{u}}\hat{\boldsymbol{\omega}} + \hat{\boldsymbol{\omega}}\hat{\mathbf{u}})/2]\mathbf{H}. \end{aligned} \quad (21)$$

This expression determining the effective elastic energy of a plane sound wave in a polarised medium is the main result of this section. While deriving relationships (21), no model assumptions were used—equation (21) is as general as equation (1) for the elastic energy of the non-polarised medium. It is important that all parameters involved in equation (21), namely $\dot{\mathbf{M}}$, $\hat{\boldsymbol{\chi}}$, $\hat{\boldsymbol{\lambda}}$ and $\hat{\mathbf{c}}^{(0)}$ (or $\hat{\mathbf{c}}$ from equation (27)) are independent material constants which can be measured (naturally all these constants are the functions of temperature and field \mathbf{H}).

The most interesting is the case of $\dot{\mathbf{M}} \parallel \mathbf{H}$ where the polarised crystal energy is a minimum with respect to the crystal orientation in space. In view of the anti-symmetry

of the tensor $\hat{\omega}$ the last terms in (19) and (21) are equal to zero and, instead of (21), we have

$$w(\hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}) = -\frac{1}{2}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) \cdot \hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + w^{(0)}(\hat{\mathbf{u}}) - \frac{1}{2}\hat{\mathbf{M}} \cdot \hat{\boldsymbol{\omega}}^2\mathbf{H} \\ + [2\pi/K(n)]\{n \cdot [\hat{\boldsymbol{\chi}}(\hat{\boldsymbol{\lambda}}\hat{\mathbf{u}} - \hat{\boldsymbol{\omega}}\mathbf{H}) + \hat{\boldsymbol{\omega}}\hat{\mathbf{M}}]\}^2 \quad (22)$$

or in an equivalent form

$$w(u_{\alpha,\beta}) = \frac{1}{2}\tilde{\mathbf{C}}_{\alpha,\beta;\mu,\nu}u_{\alpha,\beta}u_{\mu,\nu}. \quad (23)$$

The relation between $\tilde{\mathbf{C}}$ from (23) with the material tensors from (22) is given in the Appendix (equations (A1–4)).

The long-wavelength sound spectrum is directly determined by the tensor $\tilde{\mathbf{C}}$:

$$\rho\ddot{u}_\alpha = -\delta w/\delta u_\alpha = \tilde{\mathbf{C}}_{\alpha,\beta;\mu,\nu}\partial^2 u_\mu/\partial x_\beta\partial x_\nu.$$

Hence the equation for the velocities and polarisations of the three sound branches is

$$\rho v^2 u_\alpha = \tilde{\mathbf{C}}_{\alpha,\beta;\mu,\nu}n_\beta n_\nu u_\mu \quad (24)$$

where v is the velocity of sound.

So the effective elastic energy of a plane sound wave propagating in a polarised medium at $\hat{\mathbf{M}}\|\mathbf{H}$ is determined by either of the equivalent expressions (22) or (23). In comparison with a non-polarised medium, terms of $\hat{\mathbf{u}}\hat{\boldsymbol{\omega}}$ and $\hat{\boldsymbol{\omega}}\hat{\boldsymbol{\omega}}$ type appear in the expression for the elastic energy, and these terms disappear if we set $\mathbf{H} = \mathbf{M} = 0$. Moreover, since any polarised medium is at the same time piezomagnetic (or piezoelectric), in accordance with the work of Landau and Lifshitz (1982) the effective tensor of elastic moduli depends on the direction of the sound wave propagation. This effect, as well as all the terms in equations (19)–(22) containing π as a factor are due to the energy w_m of the non-homogeneous dipolar field \mathbf{h} from equation (18).

As was mentioned in § 1, the qualitative difference in the effective elastic energy (22) from (1) is determined by two independent circumstances: firstly, by the violation of rotational invariance of energy density at $\mathbf{H} \neq \mathbf{0}$; secondly, by the presence of long-range dipole forces which are not reduced by renormalisation of the elastic moduli.

To conclude this section, we rewrite the above formulae for the particular case when the uniaxial (hexagonal) crystal is in a field $\mathbf{H}\|\hat{\mathbf{M}}\|z$, where z is the symmetry axis. The coefficients in the effective elastic energy of a plane sound wave are expressed via the material tensors $\hat{\mathbf{c}}$, $\hat{\boldsymbol{\lambda}}$, $\hat{\boldsymbol{\chi}}$ and $\hat{\mathbf{M}}$ as well as via the field \mathbf{H} inside the sample. In the case under consideration the following components of the material tensors are non-zero:

$$\chi_{xx} = \chi_{yy} \equiv \chi_\perp \quad \chi_{zz} \equiv \chi_\parallel \quad c_{xz,xz} = c_{yz,yz} \equiv c_{44} \\ c_{xy,xy} \equiv c_{66} = \frac{1}{2}(c_{11} - c_{12}) \quad c_{xx,xx} = c_{yy,yy} \equiv c_{11} \quad c_{zz,zz} \equiv c_{33} \quad (25) \\ \lambda_{x,xz} = \lambda_{y,yz} \equiv \lambda \quad \lambda_{z,xx} = \lambda_{z,yy} \equiv \lambda_1 \quad \lambda_{z,zz} \equiv \lambda_2.$$

Quite naturally, all values in equation (25) are the functions of the field $\mathbf{H} = H_z \hat{z}$. In particular, if, in the case of either a paramagnet or an antiferromagnet at $\mathbf{H} \rightarrow \mathbf{0}$, $\hat{\mathbf{M}}$ also becomes zero, the tensor $\hat{\boldsymbol{\lambda}}$ can also become zero if at $\mathbf{H} = \mathbf{0}$ the magnetic symmetry of a crystal does not permit the existence of a linear piezomagnetic effect.

Provided that we bear in mind the fact that as far as elastic properties are concerned the hexagonal crystal possesses an axial symmetry, it is quite sufficient to consider the

case where the vector \mathbf{n} lies in the x - z plane. Thus, in equation (23) for the effective elastic energy of the sound wave, only the following terms are essential:

$$w(u_{\alpha,\beta}) = \frac{1}{2}\bar{c}_{11}u_{xx}^2 + \bar{c}_{13}u_{xx}u_{zz} + \frac{1}{2}\bar{c}_{33}u_{zz}^2 + \frac{1}{2}\bar{c}_{66}u_{y,x}^2 + \frac{1}{2}c_1u_{y,z}^2 - (\bar{l}_1u_{xx} \pm \bar{l}_3u_{zz})(u_{x,z} - u_{z,x}) + \frac{1}{2}\bar{c}_1u_{x,z}^2 + \frac{1}{2}\bar{c}_2u_{z,x}^2 + c_3u_{x,z}u_{z,x} + \dots \quad (26)$$

where

$$\bar{l}_1 = [2\pi/K(\mathbf{n})]\lambda_1(\dot{M} - \chi_{\perp}H)\chi_{\parallel}n_xn_z \quad \bar{l}_3 = [2\pi/K(\mathbf{n})]\lambda_3(\dot{M} - \chi_{\perp}H)\chi_{\parallel}n_xn_z \quad (27)$$

$$\bar{c}_1 = c_1 + [4\pi/K(\mathbf{n})][\chi_{\perp}\lambda - \frac{1}{2}(\dot{M} - \chi_{\perp}H)]^2n_x^2 \geq c_1$$

$$\bar{c}_2 = c_2 + [4\pi/K(\mathbf{n})][\chi_{\perp}\lambda + \frac{1}{2}(\dot{M} - \chi_{\perp}H)]^2n_x^2 \geq c_2 \quad (28)$$

$$\bar{c}_3 = c_3 - [\pi/K(\mathbf{n})](\dot{M} - \chi_{\perp}H)^2n_x^2 \leq c_3$$

$$c_1 = c_{44} - \lambda\chi_{\perp}H + \frac{1}{4}H(\dot{M} - \chi_{\perp}H)$$

$$c_2 = c_{44} + \lambda\chi_{\perp}H + \frac{1}{4}H(\dot{M} - \chi_{\perp}H) \quad (29)$$

$$c_3 = c_{44} - \frac{1}{4}H(\dot{M} - \chi_{\perp}H)$$

$$K(\mathbf{n}) = 1 + 4\pi[\chi_{\perp} + (\chi_{\parallel} - \chi_{\perp})n_z^2]. \quad (30)$$

The moduli c_1 , c_2 and c_3 correspond to the case of a uniformly deformed crystal (for details see § 3).

For a hexagonal crystal the system of equations (24) is split at an arbitrary direction \mathbf{n} into two independent systems (similarly to the case of a non-polarised crystal (Landau and Lifshitz 1965)). The first system describes the transverse sound with a polarisation $\mathbf{u} \parallel \mathbf{y}$. According to equations (26) and (24)

$$\mathbf{u} \perp \mathbf{n}, \mathbf{z}: \quad \rho v^2 = c_{66}n_x^2 + c_1n_z^2. \quad (31)$$

The second system leads to a biquadratic equation for two sound waves polarised in the x - z plane. The solution of this equation for an arbitrary \mathbf{n} is very cumbersome; therefore we restrict ourselves to the most interesting and simple cases of $\mathbf{n} \parallel \mathbf{z}$ and $\mathbf{n} \parallel \mathbf{x}$.

For a longitudinal sound,

$$\mathbf{u} \parallel \mathbf{n} \parallel \mathbf{z}: \quad \rho v_{\parallel z}^2 = \bar{c}_{33} = c_{33} + 4\pi\lambda_2^2\chi_{\parallel}^2/(1 + 4\pi\chi_{\parallel}) \quad (32)$$

$$\mathbf{u} \parallel \mathbf{n} \perp \mathbf{z}: \quad \rho v_{\perp z}^2 = c_{11}. \quad (33)$$

For transverse sound,

$$\mathbf{u} \perp \mathbf{n} \parallel \mathbf{z} \quad \rho v_{\parallel z}^2 = c_1 = c_{44} - \lambda\chi_{\perp}H + \frac{1}{4}H(\dot{M} - \chi_{\perp}H) \quad (34)$$

$$\mathbf{u} \parallel \mathbf{z} \perp \mathbf{n}: \quad \rho v_{\perp z}^2 = \bar{c}_2 = c_{44} + \lambda\chi_{\perp}H + \frac{1}{4}H(\dot{M} - \chi_{\perp}H) + (4\pi/[1 + 4\pi\chi_{\perp}])[\chi_{\perp}\lambda + \frac{1}{2}(\dot{M} - \chi_{\perp}H)]^2. \quad (35)$$

In a non-polarised medium without a piezomagnetic effect the velocities $v_{\parallel z}$ and $v_{\perp z}$ coincide (the reciprocity principle described in § 1) and are equal to c_{44}/ρ . In this case, however,

$$\rho(v_{\perp z}^2 - v_{\parallel z}^2) = 2\lambda\chi_{\perp}H + [4\pi/(1 + 4\pi\chi_{\perp})][\chi_{\perp}\lambda + \frac{1}{2}(\dot{M} - \chi_{\perp}H)]^2. \quad (36)$$

The first term on the right-hand side of equation (36) is due to the violation of rotational invariance in the field and the second term is the consequence of the non-local character

of dipole interaction. In the polarised crystal the sign of the difference $v_{t\perp} - v_{tz}$ can be either plus or minus, but in the non-polarised crystal (but with a piezomagnetic or a piezo-electric effect) this sign is always positive (Landau and Lifshitz 1982).

The effect of reciprocity principle violation for the transverse sound in magnetic field was first revealed by Melcher (1970).

3. Anomalies of the elastic properties at ferroelastic phase transitions in a polarised crystal

As was mentioned in § 1, the critical anomalies at ferroelastic phase transition are completely determined by peculiarities of the long-wavelength sound spectrum. The present section is aimed at showing the effect of the qualitative difference of equation (1) from (22) or (23) for the effective elastic energy of the sound wave on the critical behaviour of the system. For definiteness, we consider a ferroelastic phase transition in a uniaxial (hexagonal) crystal in the presence of a field $\mathbf{H} \parallel \mathbf{Z}$ where \mathbf{Z} is the symmetry axis. The results obtained are so typical that it is useless to consider other examples since all principal points remain unchanged.

Let us start from the definition of the ground state and the phase transition point. Without any loss of generality, we can choose the x axis such as to preserve the condition $m_y = 0$ in both phases. To avoid complications connected with the anisotropy of the tensor $\hat{\mathbf{N}}$ of demagnetisation coefficients and those due to existence of a thermodynamically stable domain structure in ferromagnets we assume that the sample has the shape of a thin sheet with the normal coinciding with the y axis. In this case the homogeneous part of a dipole field is zero at both sides of the critical point, and the contribution of the non-homogeneous part of a dipole field is known beforehand to be absent on determination of the homogeneous ground-state energy. Thus, for the ground-state energy density, instead of (11) we have

$$w = w_i - \dot{\mathbf{M}} \cdot \mathbf{H} \quad (37)$$

where $\mathbf{H} = \mathbf{H}_{\text{ext}} \parallel \mathbf{Z}$.

Relationships concerning the homogeneous ground state can be obtained from the formulae in § 2. For this purpose, it is quite sufficient to omit everywhere terms due to the dipole energy which is equivalent to omitting all terms containing π as a factor. For the effective elastic energy $\dot{w}(\hat{\mathbf{u}}, \hat{\omega})$ of the homogeneous state, instead of (22) we have

$$\dot{w}(\hat{\mathbf{u}}, \hat{\omega}) = -\frac{1}{2}(\hat{\lambda}\hat{\mathbf{u}} - \hat{\omega}\mathbf{H}) \cdot \hat{\chi}(\hat{\lambda}\hat{\mathbf{u}} - \hat{\omega}\mathbf{H}) + w^{(0)}(\hat{\mathbf{u}}) - \frac{1}{2}\dot{\mathbf{M}} \cdot \hat{\omega}^2\mathbf{H} \quad (38)$$

or in an equivalent form

$$\dot{w}(\hat{\mathbf{u}}, \hat{\omega}) = \frac{1}{2}\hat{\mathbf{u}} \cdot \hat{\mathbf{c}}\hat{\mathbf{u}} - \hat{\mathbf{u}} \cdot \hat{\mathbf{l}}\hat{\omega} + \frac{1}{2}\hat{\omega} \cdot \hat{\beta}\hat{\omega} \quad (39)$$

where $\hat{\mathbf{c}}$ is the tensor of elastic moduli corresponding to homogeneous deformation (i.e. neglecting the dipolar contribution (18) to the energy of deformed crystal)

$$c_{\alpha\beta,\mu\nu} = c_{\alpha\beta,\mu\nu}^{(0)} - \chi_{\gamma\delta}\lambda_{\gamma,\alpha\beta}\lambda_{\delta,\mu\nu}. \quad (40)$$

$\hat{\mathbf{l}}$ and $\hat{\beta}$ are determined by

$$l_{\alpha\beta,\bar{\mu}\bar{\nu}} = \frac{1}{2}\lambda_{\gamma,\alpha\beta}(\chi_{\mu\gamma}H_{\nu} - \chi_{\nu\gamma}H_{\mu}) \quad (41)$$

$$\begin{aligned} \beta_{\alpha\bar{\beta},\bar{\mu}\bar{\nu}} = & -\frac{1}{4}(H_{\nu}\dot{M}_{\alpha}\delta_{\beta\mu} - H_{\nu}\dot{M}_{\beta}\delta_{\alpha\mu} - H_{\mu}\dot{M}_{\alpha}\delta_{\beta\nu} + H_{\mu}\dot{M}_{\beta}\delta_{\alpha\nu} \\ & + \chi_{\alpha\mu}H_{\nu}H_{\beta} - \chi_{\beta\mu}H_{\nu}H_{\alpha} - \chi_{\alpha\nu}H_{\beta}H_{\mu} + \chi_{\beta\nu}H_{\mu}H_{\alpha}). \end{aligned} \quad (42)$$

The qualitative difference between equations (1) and (39) for the effective elastic energy is due exclusively to the violation of rotational invariance in the latter case. At $\mathbf{H} \rightarrow \mathbf{0}$, this difference is lost. Moreover, a comparison of equations (22) and (38) shows that always

$$w(\hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}) \geq \dot{w}(\hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}). \quad (43)$$

This is because the energy of the non-homogeneous dipole field is positive (equation (18)).

So equations (39)–(42) determine the energy of the uniformly deformed polarised crystal. In the case of hexagonal crystal and taking into account (25), we have

$$\begin{aligned} \dot{w}(\hat{\mathbf{u}}, \hat{\boldsymbol{\omega}}) = & \frac{1}{4}(c_{11} + c_{12})(u_{xx} + u_{yy})^2 + c_{13}u_{zz}(u_{xx} + u_{yy}) + \frac{1}{2}c_{33}u_{zz}^2 \\ & + \frac{1}{2}c_{66}[(u_{xx} - u_{yy})^2 + 4u_{xy}^2] + 2c_{44}(u_{xz}^2 + u_{yz}^2) \\ & + 2\lambda\chi_{\perp}H(u_{xz}\omega_{xz} + u_{yz}\omega_{yz}) + \frac{1}{2}H(\dot{M} - \chi_{\perp}H)(\omega_{xz}^2 + \omega_{yz}^2) \end{aligned} \quad (44)$$

or

$$\begin{aligned} \dot{w}(u_{\alpha,\beta}) = & \frac{1}{2}c_{11}(u_{xx}^2 + u_{yy}^2) + c_{12}u_{xx}u_{yy} + \frac{1}{2}c_{33}u_{zz}^2 + 2c_{66}u_{xy}^2 + c_{13}u_{zz}(u_{xx} + u_{yy}) \\ & + \frac{1}{2}c_1(u_{x,z}^2 + u_{y,z}^2) + \frac{1}{2}c_2(u_{z,x}^2 + u_{z,y}^2) + c_3(u_{x,z}u_{z,x} + u_{y,z}u_{z,y}) \end{aligned} \quad (45)$$

where the moduli c_1 , c_2 and c_3 are determined in (29).

If there are no mechanical stresses at the crystal boundary (free specimen), the hexagonal phase stability region is determined by the positive definiteness of the quadratic forms (44) and (45). Stability with respect to isomorphous distortions is determined by the inequalities

$$(c_{11} + c_{12})c_{33} - 2c_{13}^2 > 0 \quad c_{11} + c_{12}, c_{33} > 0. \quad (46)$$

Stability with respect to rhombic distortions is determined by the inequality

$$c_{66} = \frac{1}{2}(c_{11} - c_{12}) > 0. \quad (47)$$

Stability of a crystal with respect to monoclinic distortions is determined by the relationships

$$c_{44}H(\dot{M} - \chi_{\perp}H) > (\lambda\chi_{\perp}H)^2 \quad c_{44}, H > 0 \quad (48)$$

or, in an equivalent form,

$$c_1c_2 > c_3^2 \quad c_1, c_2 > 0. \quad (49)$$

It should be mentioned that under the conditions of crystal stability against spontaneous deformations, isothermal moduli should be used because similar inequalities for adiabatic moduli appear to be weaker. In this particular case, equations (47)–(49) for isothermal and adiabatic cases exactly coincide (see comment after equation (10)). For the conditions of stability against isomorphic deformations, it is isothermal moduli that should be used in equations (46).

We shall regard conditions (46) and (47) as fulfilled and consider a ferroelastic phase transition accompanied by the appearance of monoclinic distortions. This phase transition occurs by two-dimensional active irreducible representation of the hexagonal point group. This representation is realised by pairs of values

$$\{m_x, m_y\}, \{u_{xz}, u_{yz}\} \quad \text{or} \quad \{\omega_{xz}, \omega_{yz}\}. \quad (50)$$

According to (48) and (49) the phase transition point in a free specimen is determined by either of the equivalent inequalities

$$c_{44} (\dot{M} - \chi_{\perp} H) = \lambda^2 \chi_{\perp}^2 H \quad \text{or} \quad c_1 c_2 = c_3^2. \quad (51)$$

On passing the critical point (51), a free specimen undergoes a spontaneous lowering of symmetry, i.e. monoclinic distortions ($u_{xz} \neq 0$) appear and the specimen changes orientation with respect to the external field ($\omega_{xz} \neq 0$) so as to provide fulfilment of the condition $\mathbf{M} \parallel \mathbf{H} \parallel \mathbf{Z}$ in the asymmetric phase.

Comparison of (51) with (34) and (35) shows that the velocity of sound at the critical point remains finite. † In the case of the ferroelastic phase transition, this means that the abnormally developed inhomogeneous fluctuations in the critical region are absent and that it is impossible for the system to split into domains. The thermodynamic potential as well as all its derivatives with respect to temperature and field $\mathbf{H} = H_z$ remain finite and continuous on approaching the critical point from both the symmetric and the asymmetric phase sides. At the phase transition point itself, both the heat capacity and the susceptibility χ_{zz} undergo a finite jump. The Landau theory quite adequately describes a phase transition of this type.

The situation changes drastically if the field \mathbf{H} tends to zero. In this case the critical point is determined by the condition

$$c_{44} = 0 \quad (\text{at } \mathbf{H} = 0) \quad (52)$$

and two types of critical behaviour are possible.

(i) If \dot{M} or at least $\hat{\lambda}$ is non-zero, then according to (35) and (34) the transverse velocity v_{tz} of sound with $\mathbf{n} \parallel \mathbf{z}$ becomes zero at the critical point. This leads to a non-analytical character of the thermodynamic potential at $T \rightarrow T_c$; the heat capacity remains finite at the critical point, but its temperature derivative has a divergence as in the case considered by Villain (1970) and Levanuk and Sobianin (1970).

(ii) In the case of either a paramagnet or an antiferromagnet without a piezo-effect (i.e. at \dot{M} and $\hat{\lambda}$ equal to zero at $\mathbf{H} = 0$), at the critical point the velocity $v_{t\perp}$ of z -polarised sound with $\mathbf{n} \perp \mathbf{z}$ also becomes zero. The dimensionality of the space of wavevectors corresponding to anomalous fluctuations is equal to two (x - y plane) similarly to the uniaxial ferroelectric case and heat capacity has a logarithmic divergence.

In both cases in the asymmetric phase the specimen can split into plane-parallel domains (in the first case the plane of domain walls coincides with the basal plane x - y).

A completely different situation occurs when the boundary conditions are such as to provide zero displacements $\mathbf{u}(\mathbf{r})$ at the specimen surface (a 'squeezed' specimen). One can show that in this case the point of stability loss at the ferroelastic phase transition coincides with the point where the velocity of sound becomes zero even at $\mathbf{H} \neq 0$. Since the character of critical anomalies is determined by the dimensionality of the space of wavevectors corresponding to anomalous fluctuations, in this case one has to know which of the equations for the transverse velocity of sound, equation (34) or (35), is the first to become zero. If $v_{tz} < v_{t\perp}$, the character of critical anomalies is the same as in case (i) at $\mathbf{H} = 0$. If $v_{tz} > v_{t\perp}$, the critical anomalies are similar to case (ii) at $\mathbf{H} = 0$. Finally, it should be noted that, to preserve the boundary condition of the absence of displacements at the sample surface, the sample inevitably splits into compensating domains at the moment of passing the ferroelastic phase transition point.

† Thus the conclusions of Cowley (1976) and David (1983) concerning the behaviour of the elastic properties of crystals at ferroelastic phase transitions are valid only in the absence of magnetic and elastic fields.

It should be noted that, from the symmetry point of view, all the above situations are equivalent since everywhere we deal with the second-order phase transition from the same phase by the same two-dimensional irreducible representation. Nevertheless the critical behaviour of the system as a function of the boundary conditions and the relationship between the material constants appear to be essentially different.

Let us outline the problem of adequacy of magnetostatic (or electrostatic) approximation for the description of acoustic properties of polarised crystals with a linear piezomagnetic (or piezo-electric) effect. Sound propagation in these media is always accompanied by the appearance of the time-dependent polarisation and, in the general case, one cannot speak separately about sound and electromagnetic waves. It should be noted that, because of this, even at $q \rightarrow 0$ the velocity of sound is not determined by static (adiabatic) elastic moduli. However, the difference between the values obtained within the magnetostatic and the electrostatic approximation is very small, of the order of the ratio of the velocity of sound to the velocity of light. If the phase transition order parameter is transformed as the linear combination of deformation tensor components and at the same time as either the magnetic or electric polarisation (and it is this situation which was considered above), then formally we have the same reason to state that the velocity of sound becomes zero at the critical point as for the fact that the electromagnetic wave velocity becomes zero. To be more exact, we should say that the velocity of certain Goldstone waves (which actually are the coupled vibrations of elastic and electromagnetic fields) becomes zero. In this paper, we shall not go into the details of this problem. We shall mention only that taking into account the effect of electromagnetic interaction lagging does not change the most essential conclusions of the above investigation. Thus, in particular, the structure and energy of softening branches of the spectrum at $q \rightarrow 0$ and $T \rightarrow T_c$ are still determined by the 'equilibrium' equations (20) and (22); however, the velocity of the corresponding excitations will differ from the values given by equations (24) or (34) and (35) by some numerical factor which depends slightly on temperature.

Naturally, our investigation of the acoustic properties of crystals at the ferroelastic phase transition is valid only for long-wavelength sound—to be exact, for sound with $q \ll \xi^{-1}$, where ξ is the correlation length of critical fluctuations. In the most important particular case (a free sample at $H \neq 0$), anomalous critical fluctuations are totally absent, and the above formulae are valid even at the most critical point where the velocity of sound is finite.

Acknowledgment

The authors express their gratitude to Professor E P Wohlfarth for a discussion on some of the results in this paper at the conference held in 1986 in Czechoslovakia.

Appendix

The components of the tensor $\tilde{\mathbf{C}}$ from equation (23) are expressed via the material parameters $\hat{\lambda}$, $\hat{\chi}$, $\hat{\mathbf{c}}$, $\hat{\mathbf{M}}$, \mathbf{H} and the vector \mathbf{n} as

$$\hat{\mathbf{C}} = \hat{\mathbf{c}} - 2\hat{\mathbf{l}} + \hat{\beta} \quad (\text{A1})$$

where

$$\bar{c}_{\alpha\beta,\mu\nu} = c_{\alpha\beta,\mu\nu} + [4\pi/K(\mathbf{n})]\chi_{\gamma\xi}\chi_{\delta\zeta}n_{\xi}n_{\zeta}\lambda_{\gamma,\alpha\beta}\lambda_{\delta,\mu\nu}. \quad (\text{A2})$$

Here $c_{\alpha\beta,\mu\nu} = c_{\alpha\beta,\mu\nu}^{(0)} - \chi_{\gamma\delta}\lambda_{\gamma,\alpha\beta}\lambda_{\delta,\mu\nu}$ is the tensor of the elastic moduli corresponding to uniform deformation. Then

$$\bar{l}'_{\alpha\beta,\mu\nu} = \frac{1}{2}(l'_{\alpha\beta,\mu\nu} - l'_{\alpha\beta,\nu\mu}) = \bar{l}'_{\beta\alpha,\mu\nu} \quad (\text{A3})$$

$$\begin{aligned} \bar{\beta}'_{\alpha\beta,\mu\nu} = & \frac{1}{6}(\beta'_{\alpha\beta,\mu\nu} - \beta'_{\beta\alpha,\mu\nu} - \beta'_{\alpha\beta,\nu\mu} + \beta'_{\beta\alpha,\nu\mu} + \beta'_{\mu\nu,\alpha\beta} - \beta'_{\nu\mu,\alpha\beta} \\ & - \beta'_{\mu\nu,\beta\alpha} + \beta'_{\nu\mu,\beta\alpha}) \end{aligned} \quad (\text{A4})$$

where

$$l'_{\alpha\beta,\mu\nu} = \chi_{\mu\gamma}\lambda_{\gamma,\alpha\beta}H_{\nu} - [4\pi/K(\mathbf{n})]\chi_{\gamma\delta}n_{\delta}\lambda_{\gamma,\alpha\beta}(\chi_{\mu\xi}n_{\xi}H_{\nu} + n_{\nu}\dot{M}_{\mu})$$

$$\begin{aligned} \beta'_{\alpha\beta,\mu\nu} = & -H_{\nu}(\dot{M}_{\alpha}\delta_{\beta\mu} + \chi_{\alpha\mu}H_{\beta}) \\ & + [4\pi/K(\mathbf{n})](n_{\beta}n_{\nu}\dot{M}_{\alpha}\dot{M}_{\mu} + 2\chi_{\mu\gamma}n_{\gamma}n_{\beta}\dot{M}_{\alpha}H_{\nu} + \chi_{\mu\gamma}n_{\gamma}\chi_{\alpha\delta}n_{\delta}H_{\beta}H_{\nu}). \end{aligned}$$

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