Critical dynamics at ferroelastic phase transitions in an external field

V. G. Bar'yakhtar, I. M. Vitebskiĭ, N. M. Lavrinenko, and V. L. Sobolev

Physicotechnical Institute, Donetsk
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It is shown that a uniform external field qualitatively alters the pattern of critical anomalies at ferroelastic phase transitions. It is found that effects due to the breakdown of the rotational invariance of the system energy in an external field play a fundamental role. The general conclusions are illustrated for examples of spin-reorientation phase transitions in magnets.

1. INTRODUCTION

The interaction of the order parameter of a phase transition with macroscopic strains of the crystal lattice can have a decisive influence on the critical dynamics.[1,2] Even a small strictive coupling can lead to qualitative changes in the dynamics of the order parameter and to anomalies of the elastic properties of the crystals in the critical region. Striction effects are manifested most clearly at phase transitions that are classified on the basis of symmetry as proper ferroelastic transitions. In this case there is a linear relation between the order parameter and the nonisomorphic striction. If the transition is of the soft-mode type, the spectrum of oscillations of the order parameter develops a strictive gap, which does not vanish at the critical point (many examples of this kind have been analyzed in Refs. 3–5). In this case the leading anomalies are observed in the elastic properties of the system.

The present paper is devoted to the study of these anomalies at phase transitions in polarized media in external fields. The elastic anomalies discussed below are completely general in nature and do not depend on the specific physical causes leading to the proper ferroelastic phase transition. In particular, our treatment applies to an extremely broad class of magnetic spin-reorientation phase transitions, the majority of which are classified by symmetry as proper ferroelastic transitions.

Because the spectrum of acoustic vibrations of an unbounded crystal is nonactivational, for small wave vectors q the vibrations in a sound wave occur so slowly that any of the subsystems (e.g., the magnetic subsystem) has time to readjust to the strain field arising in the sound wave. It is for this reason that the sound velocity for $q \to 0$ is determined by the static elastic constants of the crystal, which include contributions from all the subsystems that interact with the strains. It is this region of the acoustic vibration spectrum that we will be studying in this paper. At large wave vectors an approach based on the continuum theory of elasticity is inadequate, and it becomes necessary to write out the explicit dynamical equations for the other (e.g., magnetic) subsystems of the crystal that interact with the sound.[6]

In the absence of magnetic and electric fields, the energy density w of a slightly strained crystal is determined by the symmetric strain tensor $u_{\alpha\beta}$ (Refs. 7 and 8)

$$w = {}^1/_2 c_{\alpha\beta,\,\mu\nu} u_{\alpha\beta} u_{\mu\nu}. \tag{1}$$

This is a consequence of the fact that w is independent of the spatial orientation of the crystal volume element (i.e., a consequence of the rotational invariance). Here and below, the components $u_{\alpha\beta}$ of the strain tensor describe the small deviations from the equilibrium position that arise in the sound wave. The same is true for the remaining components of the distortion tensor considered below:

$$u_{\alpha,\,\beta} \equiv \partial u_\alpha / \partial x_\beta = u_{\alpha\beta} + \omega_{\alpha\beta}, \tag{2}$$

where

$$u_{\alpha\beta} = {}^1/_2 (u_{\alpha,\,\beta} + u_{\beta,\,\alpha}), \quad \omega_{\alpha\beta} = {}^1/_2 (u_{\alpha,\,\beta} - u_{\beta,\,\alpha}).$$

At the point of a proper ferroelastic transition, the quadratic form (1) suffers a loss of positive definiteness. Here the anomalies associated with critical behavior are completely determined by the anomalies of the elastic properties of the crystal.

In fact, in the absence of external fields, the expansion of the free energy in powers of the order parameter η and strain tensor $\hat{u}$ has the following structure for a ferroelastic phase transition:

$$w(\eta,\ \hat{u}) = \mathring{w}(\eta) + \mathring{w}(\hat{u}) - \lambda_{i,\,\mu\nu} \eta_i u_{\mu\nu}. \tag{3}$$

Here

$$\mathring{w}(\eta) = {}^1/_2 \mathring{a} \eta_i \eta_i + \ldots, \quad \mathring{w}(\hat{u}) = {}^1/_2 \mathring{c}_{\alpha\beta,\,\mu\nu} u_{\alpha\beta} u_{\mu\nu}, \tag{4}$$

η_i are the components of the order parameter (the index i is the number of the row of the irreducible representation responsible for the phase transition), and $\mathring{c}_{\alpha\beta,\mu\nu}$ are the bare elastic constants.

Let us transform from the ordinary Cartesian components $u_{\alpha\beta}$ of the strain tensor to their linear combinations which form the irreducible representations of the symmetry group of the high-symmetry phase

$$u_{\mu\nu} = \xi^{(\Gamma)}_{i,\,\mu\nu} u^{(\Gamma)}_i, \tag{5}$$

here Γ is the number of the irreducible representation, and i is the row index in it. Substituting (5) into (3) and (4), we get

$$w(\eta,\ \hat{u}) = \mathring{w}(\eta) + \mathring{w}(\hat{u}) - \lambda \eta_i u_i; \quad \mathring{w}(\hat{u}) = {}^1/_2 \mathring{c} u_i u_i + \ldots. \tag{6}$$

Here we have written out in $\mathring{w}(\hat{u})$ only the term corresponding to the irreducible representation according to which the order parameter transforms ($u_i \propto \eta_i$). We shall drop the index Γ for this representation. The quantity $\mathring{c}$ is the corre-

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sponding linear combination of the elastic constants $\mathring{c}_{\alpha\beta,\mu\nu}$.

Using the condition $\partial w/\partial\eta_i = 0$, we eliminate the variables η_i from (6). As a result, we arrive at expression (1) for the total energy

$$w(\hat{u}) = {}^1/_2 c_{\alpha\beta,\mu\nu} u_{\alpha\beta} u_{\mu\nu} \equiv {}^1/_2 c u_i u_i + \ldots, \quad (7)$$

where

$$c = \mathring{c} - \lambda^2/\mathring{a}, \quad \eta_i = \lambda u_i/\mathring{a}. \quad (8)$$

Eliminating the components of the strain tensor from Eq. (6), we get

$$w(\eta) = {}^1/_2 a \eta_i \eta_i + \ldots, \quad (9)$$

where

$$a = \mathring{a} - \lambda^2/\mathring{c}, \quad u_i = \lambda \eta_i/\mathring{c}. \quad (10)$$

The stability region of the symmetric phase is determined by one of the equivalent inequalities

$$c \geqslant 0, \quad a \geqslant 0. \quad (11)$$

At the point of the ferroelastic transition we have

$$c = 0 \quad (a = 0) \quad (12)$$

and quadric forms (1), (3), (6), (7), and (9) simultaneously suffer a loss of positive definiteness.

The critical anomalies of the elastic properties, including the critical behavior of long-wavelength sound, can be analyzed by proceeding from the equivalent expressions (1) or (7) for the true (renormalized) elastic energy. We recall that these expressions are valid only in the absence of external fields, since they were derived using the invariance of the energy density with respect to the spatial orientation of the crystal volume element. In the presence of external fields, even uniform ones, this invariance is either partially or totally (in the case of crossed electric and magnetic fields) lost. Therefore, before turning to a discussion of the effects due to the breakdown of the rotational invariance, let us recall the acoustic anomalies that arise at proper ferroelastic transitions in the absence of external fields.

Analysis of the stability (positive definiteness) of quadratic form (1) or (7) together with the expressions for the velocity of long-wavelength sound in crystals of different systems implies that in all cases

1) The velocity of one or two branches of transverse sound, propagating in definite directions, goes to zero at the critical point (12).

Further, a direct consequence of (1) is the so-called reciprocity principle, which holds that the velocity of transverse sound having x polarization and propagating in the y direction is equal to that of transverse sound having y polarization and propagating in the x direction (x and y are two mutually perpendicular directions). Thus, in addition to assertion 1), it is also true that

2) If the velocity of an x polarized wave with $\mathbf{q}\|y$ goes to zero, then so must the velocity of a y polarized wave with $\mathbf{q}\|x$.

In the case considered above, the critical anomalies of the dynamics are uniquely determined by the symmetry of the initial phase and the transformation properties of the order parameter. In fact, the macroscopic symmetry of the initial phase and the index of the irreducible representation responsible for the ferroelastic phase transition uniquely determine the anomalies of the elastic properties (and thus of the long-wavelength sound) in the critical region. Moreover, it is precisely the elastic subsystem that is anomalously fluctuating in this case. Such a situation is typical in the theory of phase transitions.

However, for ferroelastic phase transitions in polarized media (in the presence of a magnetic or electric field) this conclusion is no longer valid. We shall see that at a second-order phase transition in a polarized medium the critical dynamics can differ qualitatively from one case to another even if the order parameter has the same symmetry. This is due to the breakdown of the rotational invariance of the energy density w in the presence of a field.

As we have mentioned, acoustic vibrations in the limit of small wave vectors $\mathbf{q}$ can always be treated in the continuum theory of elasticity, and here the sound velocity will be determined by the same parameters (elastic constants) as are the static elastic properties. However, when a field is present, expression (1) for the energy density no longer applies—its derivation[7] was based on rotational invariance, the breakdown of which gives rise to an antisymmetric part $\omega_{\alpha\beta}$ of the distortion tensor $u_{\alpha,\beta}$ in the expression for w. The influence of these additional terms in w on the critical dynamics at the point of a proper ferroelastic phase transition will be considered in the following section.

Finally, we note that a breakdown of the reciprocity principle for sound can originate not only in a breakdown of the rotational invariance in an external field but also in the influence of the long-range dipole interaction. This second mechanism cannot be taken into account in the continuum theory of elasticity because of the long-range character of dipole interactions.

2. INFLUENCE OF A FIELD ON THE ANOMALIES OF THE ELASTIC PROPERTIES AT A FERROELASTIC PHASE TRANSITION

For the sake of definiteness, let us consider a uniaxial (hexagonal) crystal in a field $\mathbf{H}\|z$, where z is the symmetry axis. The portion of the elastic energy that has complete rotational invariance is[7]

$$w(H=0) = {}^1/_2 c_{11}(u_{xx}^2 + u_{yy}^2) + c_{12} u_{xx} u_{yy} + 2c_{66} u_{xy}^2 + c_{13}(u_{xx} u_{zz} + u_{yy} u_{zz}) + {}^1/_2 c_{33} u_{zz}^2 + 2c_{44}(u_{xz}^2 + u_{yz}^2). \quad (13)$$

A field $\mathbf{H}\|z$ leaves only axial (one-parameter) rotational invariance. Here the symmetry admits the existence of the following contributions to w [in addition to those in (13)]

$$2\lambda(u_{xz}\omega_{xz} + u_{yz}\omega_{yz}) + 2\beta(\omega_{xz}^2 + \omega_{yz}^2). \quad (14)$$

Naturally, for $\mathbf{H} = 0$ we have $\lambda = \beta = 0$.

With allowance for (13) and (14), we have for the elastic energy at $\mathbf{H} \neq 0$

$$w = {}^1/_2 c_{11} u_{xx}^2 + c_{13} u_{xx} u_{zz} + {}^1/_2 c_{33} u_{zz}^2 + 2c_{44} u_{xz}^2$$

$$+2\lambda u_{xz}\omega_{xz}+2\beta\omega_{xz}^2+\dots \quad (15)$$

or, equivalently,

$$w={}^1/_2c_{11}u_{xx}^2+c_{13}u_{xx}u_{zz}+{}^1/_2c_{33}u_{zz}^2+{}^1/_2c_1u_{x,z}^2+{}^1/_2c_2u_{z,x}^2+c_3u_{x,z}u_{z,x}+\dots, \quad (16)$$

where

$$c_1=c_{44}+\beta+\lambda,\ c_2=c_{44}+\beta-\lambda,\ c_3=c_{44}-\beta. \quad (17)$$

In (15) and (16) we have not written out the terms containing the y coordinate; they can be obtained from the terms shown by letting $x\to y$.

In view of the symmetry of the problem, it is sufficient to consider the case in which the wave vector $\mathbf{q}$ lies in the xz plane. The velocity of sound polarized in the xz plane is given by the solution of the biquadratic equation

$$(\rho v^2)^2-\rho v^2\{c_1+c_{33}+(c_{11}-c_{33}-c_1+c_2)\varkappa\}+\{c_1+(c_{11}-c_1)\varkappa\}\{c_{33}+(c_2-c_{33})\varkappa\}-d^2\varkappa(1-\varkappa)=0, \quad (18)$$

where

$$\varkappa=\sin^2\theta=q_x^2q^{-2},\ d=c_{13}+c_3. \quad (19)$$

For $\mathbf{q}\|z$ and $\mathbf{q}\|x$ the two acoustic branches described by (18) correspond to longitudinal and transverse sound:

$$\rho v_{t_1}^2=c_1,\ \rho v_l^2=c_{33}\ \text{for}\ q\|z\ (\varkappa=0), \quad (20)$$

$$\rho v_{t_2}^2=c_2,\ \rho v_l^2=c_{11}\ \text{for}\ q\|x\ (\varkappa=1). \quad (21)$$

The breakdown of the reciprocity principle in a field $\mathbf{H}\|z$ reduces to a difference in the values of v_{t_1} and v_{t_2}:

$$\rho(v_{t_1}^2-v_{t_2}^2)=c_1-c_2=2\lambda. \quad (22)$$

The third branch of the acoustic spectrum describes transverse sound with y polarization:

$$\rho v_t^2=c_{66}\varkappa+c_1(1-\varkappa). \quad (23)$$

Let us study the anomalies of the acoustic spectrum at the point of a proper ferroelastic phase transition accompanied by the appearance of monoclinic distortions. Such a transition occurs according to a two-dimensional irreducible representation of the symmetry group of the initial phase. This representation is formed by the pairs $\{u_{xz},u_{yz}\}$ or $\{\omega_{xz},\omega_{yz}\}$.

If the boundary conditions are such that there is no stress at the surfaces of the sample (the sample is free), then the stability region of the symmetric phases is the region in which quadratic form (15) or (16) is positive definite.

Stability against isomorphic strains is ensured by the inequalities

$$c_{11}c_{33}-c_{13}^2>0,\quad c_{11},\ c_{33}>0, \quad (24)$$

which we assume are satisfied.

The conditions for stability against monoclinic distortions are

$$\beta c_{44}>{}^1/_4\lambda^2,\quad \beta,\ c_{44}>0, \quad (25)$$

or, equivalently,

$$c_1c_2>c_3^2,\quad c_1,\ c_2>0. \quad (26)$$

The critical point of the proper ferroelastic transition is determined by the condition

$$4\beta c_{44}=\lambda^2\ \text{or}\ \ c_1c_2=c_3^2, \quad (27)$$

where β, c_{44}, c_1 and c_2 are positive (for $\mathbf{H}\neq 0$).

On passage through critical point (27) in a free sample there is a spontaneous lowering of the symmetry: monoclinic distortions arise. Here the sound velocity remains finite. A distinctive feature of such a phase transition is the absence of anomalously large inhomogeneous fluctuations in the critical region, and also the impossiblity of breaking the sample up into domains. Here Landau theory is fully adequate to describe the critical behavior of such a system.

It is easy to see that for $\mathbf{H}=0$ the situation becomes fundamentally different. Here $\lambda=\beta=0$, $c_1=c_2=c_3\equiv c_{44}$, and the critical point is determined by the condition $c_{44}=0$. At this point, according to (20) and (21), we have $v_{t_1}=v_{t_2}=0$, and the anomalous fluctuations of the order parameter that are characteristic for ferroelastic transitions[1,2] are observed in the system, and here the sample can be broken up into domains.

The situation will be different if the boundary conditions are such that the displacements $\mathbf{u}(\mathbf{r})$ are equal to zero on the surface of the sample (a clamped sample): $\mathbf{u}(\mathbf{r})|_s=0$, where S is the surface of the sample. For example, a sample in the form of a thin slab with normal $\mathbf{n}\|z$ might be cemented to a backing plate. In such a case the point at which the elastic subsystem loses stability coincides with the point at which the sound velocity goes to zero. More precisely, under the condition $c_{11}c_{33}>d^2$ the following transverse-sound branches go to zero at the critical point:

$$\mathbf{q}\|z,\ \mathbf{u}\perp z\ \text{for}\ \lambda<0\ (2\ \text{branches}, \quad (28)$$

$$\mathbf{q}\perp z,\ \mathbf{u}\|z\ \text{for}\ \lambda>0\ (1\ \text{branch}, \quad (29)$$

where $\mathbf{u}$ is the polarization vector of the transverse sound. Importantly, from the symmetry standpoint situations (28) and (29) are equivalent: in both cases there is a second-order phase transition, which occurs from the same phase acording to the same representation. Nevertheless, the critical behavior of the system in cases (28) and (29), and also in case (27) (for the free boundary conditions), is substantially different.

Here we analyze only one example of a proper ferroelastic transition in a system with broken rotational invariance. The aforementioned features due to the presence of an external field turn out to be so typical that it is pointless to give other examples—all the fundamental features remain unchanged.

In the next section we illustrate the results for the simplest example.

3. SPIN-REORIENTATION PHASE TRANSITIONS IN A FIELD

Let us study the features of the elastic properties (sound) in an easy-plane ferromagnet near the point of a spin-reorientation phase transition in a field $\mathbf{H}\|z$. This transition involves the appearance of a transverse (to the z axis) component of the magnetization vector $\mathbf{m}$ as the field decreases. Simultaneously, the strictive coupling gives rise to u_{xz} or u_{yz} components of the distortion tensor. According to a symmetry classification, this transition is a proper ferroelastic transition, and all of the discussion of the previous section carries over to this case.

Without loss of generality, we can assume that the spin reorientation occurs in the xz plane. Then, for studying the stability region of the symmetric phase (with $\mathbf{m}\|\mathbf{H}\|z$) one can start from the following expression for the energy density w, in which we have kept only the terms which are quadratic in the small deviations from the symmetric phase:

$$w=w_{\mu}+w_{\mu y}+w_{y}, \tag{30}$$

where

$$w_{\mu}=-{}^1/_2K(m_x-m_0\omega_{xz})^2-Hm_z+\dots, \tag{31}$$

$$w_{\mu y}=2B_{44}m_0(m_x-m_0\omega_{xz})u_{xz}+\dots, \tag{32}$$

$$w_y={}^1/_2c_{11}u_{xx}^2+{}^1/_2c_{33}u_{zz}^2+c_{13}u_{xx}u_{zz}+2c_{44}^{(0)}u_{xz}^2+\dots. \tag{33}$$

All the contributions to w except the Zeeman term have complete rotational invariance to second order in small deviations from the symmetric phase. It is the Zeeman term Hm_z in (31) that is responsible for the breakdown of the rotational invariance.

Using the condition that w from (3) be minimum with respect to the direction of the vector $\mathbf{m}$, we eliminate the spin variables from w. As a result, the expression for the energy density assumes the form in (15), with

$$c_{44}=c_{44}^{(0)}-\frac{B_{44}^2m_0^3}{H-Km_0},\quad \lambda=-\frac{Hm_0^2B_{44}}{H-Km_0},$$
$$\beta=-\frac{Hm_0^2K}{4(H-Km_0)}. \tag{34}$$

All the formulas and arguments of the previous section remain in force. From a comparison of (22) and (34), we see that the nonreciprocity in this case is linear in the field and is determined by the magnetostriction constant B_{44}.

This effect was first pointed out by Melcher.[9] However, in Ref. 9 and in a number of subsequent papers, the possibility of describing this effect was linked to the use of the theory of finite deformations. From the previous discussion it is obvious that here we are not dealing with finite deformations, but with the linear theory of elasticity, although for systems with broken rotational invariance.

Finally, we note that in the present model of an easy-plane ferromagnet with free boundary conditions, the phase with $\mathbf{m}\|\mathbf{H}\|z$ is unstable, since at any nonzero field it is favorable for the sample to turn in such a way that the external field lies in the basal plane.

Up till now we have ignored the contribution of the magnetic dipole interaction to the system energy. Since this is a long-range interaction, allowance for it does not reduce to a renormalization of the constants in (15) and (16). In particular, the shape of the sample becomes important. If the sample is a slab with normal $\mathbf{n}\|\mathbf{H}\|z$, then, following Ref. 6, we easily see that allowance for the dipole interaction reduces to the replacement of the external field H in (34) by $H_i=4\pi m_0\varkappa$, where $H_i=H-4\pi m_0$ is the magnetic field within the slab. Here, in place of (22), we get for the difference in the velocities of transverse sound

$$v_{t_1}^2-v_{t_2}^2=-\frac{B_{44}m_0^2H_i}{\rho(H_i-Km_0)}-\frac{\pi m_0^4(B_{44}-K)^2}{\rho(H_i-Km_0)(H_i-Km_0+4\pi m_0)}. \tag{35}$$

The first term on the right-hand side of (35) is due to the breakdown of the rotational invariance of the energy density w in a field H_i, while the second term is of a purely dipolar origin and is not related to the breakdown of rotational invariance. The contribution of this term to the velocity difference (35) is always negative, whereas the contribution from the first term can, in principle, be of either sign.

Expressions analogous to (35) for antiferromagnets in a magnetic field are given in Ref. 9 and 10.

Naturally, expressions (34) can also be obtained from the solution of dynamical equations for coupled magnetoelastic waves (see Ref. 10 and the references cited therein). However, from the arguments given above we see that if one is interested only in long-wavelength sound there is no point in writing out these equations of motion. In regard to the spin dynamics in the critical region, these questions have already been studied in detail (see, e.g., Refs. 3 and 10). In particular, it is well known that at the critical point of a proper ferroelastic phase transition, the activational gap in the magnon spectrum does not vanish, and the role of the anomalously fluctuating system (if there is one) is taken over by the sound.

The last section of this paper is strictly for illustration: all the fundamental results are contained in Sec. 2. Nevertheless, we have found it necessary to give the computations here for the simplest model example in order to distinguish clearly the fundamental role of the effects of broken rotational invariance at ferroelastic phase transitions.

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