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Dynamics of the interface in piezoelectric with the temperature gradient under cooling with a finite rate

© A.A. Chevrychkina, A.L. Korzhenevskii

Institute of Problems of Mechanical Engineering, Russian Academy of Sciences,
St. Petersburg, Russia

E-mail: alekorzh@mail.ru

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The paper analyzes the dynamics of an interphase boundary (IPB) entrained by a moving isotherm with a given velocity $V_{iso}(t)$ in a ferroelectric with a piezoelectric effect in the paraphase. Within the framework of the model, in which the dependence of the thermodynamic potential on polarization is given by two intersecting parabolas, a non-linear differential equation of the second order is derived for the $X(t)$ coordinate of the nonstationary IFB. It is shown that this equation describes the dynamics of the IFB well for almost all experimentally used modes of isotherm motion.

Keywords: Phase transition, piezoelectric effect in paraphase, temperature gradient, moving isotherm, derivation of the differential equation of motion for nonstationary interphase boundary.

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1. Introduction

The experimental study of the phase transition (PT) kinetics is in many cases complicated by the appearance of a huge number of growing nuclei of a new phase, often with a complex morphology that critically depends on the given external conditions. The theoretical describing of the PT macrokinetics entails the need for an averaged statistical description of the dynamics of the intermediate two-phase state of the material, which in the general case seems to be an extremely difficult task. Calculations are somewhat simplified if the problem of the evolution of the state in the entire volume of the sample can be reduced to the solution of the problem of the interphase boundaries (IPB) movement, see, for example, [1,2] and the literature cited therein. Real analytical calculations become possible, however, only in the case of a small number of nuclei, which means that the deviation of the homogeneous state of the material from the equilibrium state should be small throughout the entire PT process. Accordingly, the velocities of IPB of nuclei turn out to be relatively low, so the complete PT process is necessarily greatly extended in time, which is not always convenient. Also, it is clear that a theoretical description of the kinetics of „fast" PTs occurring under highly nonequilibrium conditions becomes unattainable.

Although, despite the above-mentioned difficulties in interpreting the observational results, most experiments on the study of PT kinetics are traditionally still carried out under homogeneous external conditions, combinations of controlled spatially inhomogeneous and time-varying fields under some solid-state PTs were used to specifically study the dynamics of isolated IPBs. In particular, this technique was used to study the dynamics of single IPBs in $BaTiO_3$, $PbTiO_3$ ferroelectrics and in $NaNbO_3$ antiferroelectric material [3–9]. Apart from structural PTs, an experimental scheme with a given temperature gradient and a fixed isotherm velocity was used to study the so-called „massive transformations" [10,11]. In addition, it is widely used for growing crystals from melts and solutions [12], as well as during the so-called directional crystallization of alloys [13].

It is worth noting that the desire to produce the most homogeneous material-product results in the situation that the simplest version of this combination is used in these experiments: a constant temperature gradient ∇T is created in the sample, with cooling or heating rate of the sample $\dot{T}$ being constant over time. From the theoretical point of view, this means that when mathematically analyzing possible solutions to nonlinear equations of PT kinetics, it is sufficient to limit the analysis to the search for stable self-similar IPBs moving at the same constant velocity $V_{iso} = -\dot{T}/\nabla T$ as that of the PT temperature isotherm $T = T_c$. At the same time, the question remains open of how to describe the dynamics of the IPB in non-stationary modes, when the experimentally specified isotherm velocity is not constant, $V_{iso} = V_{iso}(t)$.

The geometric movement of the IPB is accompanied not only by a change in the phase state of the material but also by a change in the values of those internal degrees of freedom that are quite strongly „coupled" with the field of the order parameter (OP). Due to the fact that the relaxation times of these degrees of freedom are finite, their response to a change in phase state is delayed and depends, generally speaking, on the trajectory of the IPB at all previous times. In turn, the degree of this reaction affects the movement of the IPB in subsequent times. Physically this happens

because the internal degrees of freedom find themselves in the vicinity of the moving IPB in a nonequilibrium state, and this leads to energy dissipation and generates „friction forces“, as well as „inertia and hyperinertia forces“, effectively acting on the IPB [2,14–21].

Thus, the calculation of the IPB dynamics in nonstationary external conditions is reduced to the calculation of these effective forces. Of course, their specific form depends on the actual nature of the nonstationarity. In this report, we will show, using the example of analyzing the dynamics of an IPB in a uniaxial piezoelectric undergoing a 1-st-order phase transition, how one can derive the equation of motion of a flat IPB in differential form, applicable for calculations under unsteady conditions. The importance of taking into account the effect of piezoelectric coupling on the dynamics of the IPB is due, in addition to its purely scientific significance, also to the fact that the value of the effective piezoelectric coefficient in real inhomogeneous materials increases sharply near the PT point (see, for example, [23]), which is widely used for practical purposes [24–28].

2. Statement of the problem and description of the model

Let us consider an experiment where a uniaxial ferroelectric crystal, which has a piezoelectric effect in the paraphase, is in the field of a given constant temperature gradient ∇T, directed along x axis, so that in (y, z) plane of the sample a flat IPB is formed, which coordinate in equilibrium corresponds to the position of the PT temperature isotherm $T = T_c$. At the time instant of $t = 0$, the sample as a whole begins to cool at a time-dependent rate $\dot{T}(t)$. Accordingly, the coordinate of the isotherm with $T = T_c$ will move across the crystal at a velocity of $V_{iso}(t) = -\dot{T}/\nabla T$, dragging the IPB along with it. The task is to derive an equation for the IPB coordinate $X(t)$, taking into account not only the action of thermodynamic forces on the boundary but also the action of resulting viscous stresses.

To carry out *analytically* all the necessary calculations, we use the following simplified model. First, we will consider the dissipative forces associated with heat conduction processes to be small compared to the forces caused by internal friction processes. We will take the latter into account within the dissipative function R normalized to unit area for an isotropic solid body [29]:

$$R = \int dx \left[\eta \left(v_{ik} - \frac{1}{3}\delta_{ik} v_{ll} \right)^2 + \frac{\varsigma}{2} v_{ll}^2 \right], \qquad (1)$$

where

$$v_{ik} \equiv \frac{1}{2}\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} \right), \quad v_i = \dot{u}_i,$$

u_i is the i-th component of the displacement field, η and ς are viscosity coefficients.

Second, we will represent the contribution $W(P)$ to the density of the thermodynamic potential of the crystal, associated exclusively with the magnitude of the polarization vector $P \equiv (0, 0, P_z)$, in the form of 2 intersecting parabolas [30]:

$$W(P) = \min \left[\frac{E}{2} P^2, \frac{E}{2} (P - P_S)^2 \right], \qquad (2)$$

where parameter E describes the height of the energy barrier between two coexisting phases, and P_S denotes the temperature-dependent spontaneous polarization in the polar phase. We will consider the contribution to the potential density of an elastically deformed crystal to be isotropic, and the contribution from the connection of the elastic strain field ε_{ik} with the polarization P will be written as $-dP_z\varepsilon_{xy}$.

The parameters E and P_S in (2) are related to the parameters of the more familiar PT model, which uses the Landau expansion for the thermodynamic potential density Φ in powers of OP:

$$\Phi = \frac{1}{2}\alpha_0 (T - T_0) P^2 + \frac{1}{4} B P^4 + \frac{1}{6} D P^6 + \frac{1}{2} g \left(\frac{dP}{dx} \right)^2 \qquad (3)$$

by the following relationships:

$$E = \frac{1}{2}\alpha_0 (T_c - T_0) P_*^2, \quad P_*^2 = -\frac{3B}{4D}, \quad T_c - T_0 = \frac{3}{16}\frac{B^2}{\alpha_0 D},$$

$$P_S^2 = \frac{-B}{2D}\left[1 + \sqrt{1 - \frac{4\alpha_0 D}{B^2}(T - T_0)} \right]. \qquad (4)$$

We will consider the OP dynamics to be of purely relaxation nature with a characteristic time τ.

3. Equations for the motion in volume and determining the interphase boundary position

The advantage of the model „of two parabolas“ formulated above for carrying out analytical calculations is that its dynamics are described by a system of quasi-linear equations. Taking into account the fact that we need to calculate the motion of a flat IPB lying in (z, y) plane along the $X(t)$, derivatives with respect to coordinates y, z are equal to zero and the equations of motion for polarization $P \equiv P_z$ and the displacement field induced by it u_x, u_y, u_z take on the following simple form:

$$\tau \dot{P} = E \left[-P + H\big(X(t) - x\big) + g P''_{xx} + \frac{d}{2}\frac{\partial u_y}{\partial x} \right],$$

$$\rho \ddot{u}_y = \mu \frac{\partial^2 u_y}{\partial x^2} - d\frac{\partial P}{\partial x} + \eta \frac{\partial^2 \dot{u}_y}{\partial x^2},$$

$$\rho \ddot{u}_z = \mu \frac{\partial^2 u_z}{\partial x^2} + \eta \frac{\partial^2 \dot{u}_z}{\partial x^2},$$

$$\rho\ddot{u}_x = (\lambda + 2\mu)\frac{\partial^2 u_x}{\partial x^2} + \left(\varsigma + \frac{4}{3}\eta\right)\frac{\partial^2 \dot{u}_x}{\partial x^2}. \quad (5)$$

In (5) $H(x)$ is Heaviside function, which appears in the first equation (5) after differentiating the contribution $W(P)$ (2) and plays the role of a „source". It should be noted that the directing the flat IPB motion along x axis allows getting rid of the depolarizing field and the problem of polydomainization of the sample associated with this field.

To obtain a solution to system (3) in an explicit form, it must be supplemented with the information about the IPB coordinate $X(t)$. Let us set its position by a natural condition, first used in [30]:

$$P\big(x = X(t)\big) = \frac{1}{2}P_S\big(T = T_{int}(t)\big), \quad (6)$$

where $T_{int}(t)$ is temperature at the IPB. In fact, (6) is a definition of what we consider to be the „center" of the IPB, where the dimensionless value of the order parameter is chosen to be equal to 1/2, i. e. it is „equidistant" from the values of 0 and 1 corresponding to the values of dimensionless polarization in the paraelectric phase and ferroelectric phase.

It is assumed that the temperature dependence of polarization near the transition point can be considered linear: $P_S(T \approx T_c) = P_S(T_c) + a(T - T_c)$, $a < 0$ (because OP increases with decreasing temperature). In the following, the solutions to system (5) in dimensionless variables will be analyzed, in which the new time is measured in τ, the new length is measured in $\sqrt{g}$, energy density is measured in E, polarization is measured in $P_S(T_c) = P_*$.

4. Solving the system of equations (5) and deriving the equation of the interphase boundary motion in differential form

Due to the fact that system (5) is quasilinear, it is convenient to use the Fourier transform to solve it:

$$F(k,\omega) = \frac{1}{2\pi}\int\limits_{-\infty}^{\infty}\int\limits_{-\infty}^{\infty} dx dt F(x,t)\exp[-ikx - i\omega t]. \quad (7)$$

This yields a system of algebraic equations with nonzero elements of its 4×4 matrix L_{ij} equal to

$$L_{pp} = 1 + i\omega + k^2, \quad L_{pu_y} = -\frac{ikd}{2}, \quad L_{u_y p} = ikd,$$

$$L_{u_y u_y} = L_{u_z u_z} = -\frac{1}{c_t^2}\omega^2 + k^2 + k^2 i\omega\eta,$$

$$L_{u_x u_x} = -\frac{1}{c_t^2}\omega^2 + \frac{c_l^2}{c_t^2}k^2 + k^2 i\omega\left(\varsigma + \frac{4}{3}\eta\right).$$

By inverting the matrix L_{ij}, the Fourier transforms of the polarization $P(k,\omega)$ and the non-zero component of the displacement field $u_y(k,\omega)$ can be written in the following form:

$$P(k,\omega) = H(k,\omega)\frac{Q(\omega,k)}{S(\omega,k)}, \quad (8)$$

$$u_y(k,\omega) = H(k,\omega)\frac{-ikd}{S(\omega,k)}, \quad (9)$$

where $H(k,\omega)$ is Fourier transform of the Heaviside function:

$$H(k\omega) = \frac{1}{(2\pi)^2}\int\limits_{-\infty}^{\infty} dt' \int\limits_{-\infty}^{X(t')} dx' \exp(-ikx' - i\omega t'), \quad (10)$$

$$Q(\omega,k) = -\frac{1}{c_t^2}\omega^2 + k^2 + k^2 i\omega\eta,$$

$$S(\omega,k) = (i\omega + 1 + k^2)\left(-\frac{1}{c_t^2}\omega^2 + k^2 + k^2 i\omega\eta\right) - \frac{k^2 d^2}{2}. \quad (11)$$

From (8)−(10) an expression for $P(x,t)$ can be derived through the inverse Fourier transformation:

$$P(x,t) = \frac{1}{(2\pi)^2}\int\limits_{-\infty}^{\infty} dk \int\limits_{-\infty}^{\infty} d\omega \int\limits_{-\infty}^{+\infty} dt' \int\limits_{-\infty}^{X(t')} dx'$$

$$\times \frac{Q(\omega,k)}{S(\omega,k)}\exp\big[-ikx' - i\omega t'\big]\exp\big[ikx + i\omega t\big]. \quad (12)$$

To write the equation of IPB motion in dimensionless variables using condition (4), it is necessary to substitute the dimensionless polarization value (12) taken at the boundary $x = X(t)$ into its left side, and equate the right side to 1/2. At the same time, it should be taken into account that, due to the dissipation, the coordinate of the moving IPB $X(t)$ lags behind the position of the PT isotherm T_c by a distance of h_0, which under conditions of a non-zero temperature gradient ∇T results in an additional to (12) contribution to the polarization at the boundary, which is equal to

$$a\big[T(X) - T_c\big] = a\big[\nabla T X(t) + h_0\big]. \quad (13)$$

For further calculations in expression (12) with $x = X(t)$ it is convenient to replace the integration variable $x' \to x' - X(t')$ and rewrite (12) in the following form:

$$P\big(x = X(t)\big) = \frac{1}{(2\pi)^2}\int\limits_{-\infty}^{\infty} dk \int\limits_{-\infty}^{\infty} d\omega \int\limits_{-\infty}^{+\infty} dt' \int\limits_{-\infty}^{0} dx'$$

$$\times \frac{Q(\omega,k)}{S(\omega,k)}\exp\big[-ikx' - i\omega t'\big]\exp\big[ik\big(X(t) - X(t')\big)\big]. \quad (14)$$

Then the difference $X(t) - X(t')$ in (14) can be represented in the form of a Taylor's series:

$$X(t) - X(t') = \dot{X}(t)(t - t') - \frac{1}{2}\ddot{X}(t)(t - t')^2 + \ldots \quad (15)$$

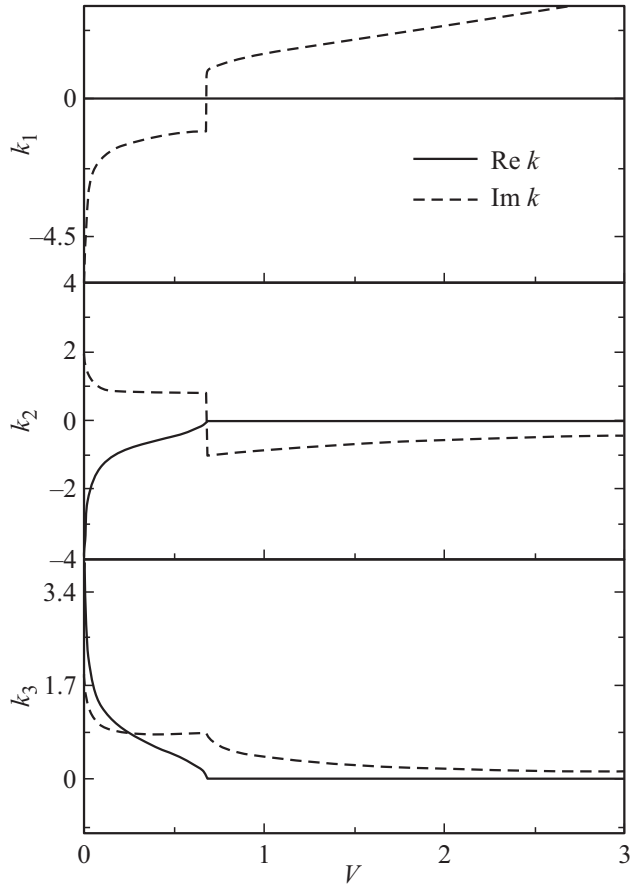

Figure 1. Dependence of the roots of $S_1(V, k)$ as a function of velocity for the following parameters: $\eta = 100$, $d = 10$, $c_t = 1400$.

Using (15), the integral over t' in (14) can be calculated term by term for an arbitrary and yet unknown IPB trajectory $X(t)$, because derivatives in (15) are taken at the moment of time t and do not depend on the integration variable t'. As a result, the right side of equation (14) can be written as a series, where $V \equiv \dot{X}(t)$:

$$f(V) + \sum_{n=1}^{\infty} C_n \partial_t^n V. \tag{16}$$

From (14)–(16) an expression for $f(V)$ can be derived:

$$f(V) = \frac{1}{(2\pi)^2} \int\limits_{-\infty}^{\infty} dk \int\limits_{-\infty}^{\infty} d\omega \int\limits_{-\infty}^{+\infty} dt' \int\limits_{-\infty}^{0} dx' \frac{Q(\omega, k)}{S(\omega, k)}$$

$$\times \exp\left[-ikx' + i\omega(t - t')\right] \exp\left[ik\left(V(t - t')\right)\right]. \tag{17}$$

It can be simplified, because the integral over t' in (17) is proportional to the delta function $\delta(kV + \omega)$; taking this into account, the subsequent integration over ω turns out to be trivial, and in the remaining integrand $\omega = -kV$ should be assumed:

$$f(V) = \frac{1}{2\pi} \int\limits_{-\infty}^{\infty} dk \int\limits_{-\infty}^{0} dx' \frac{Q(-kV, k)}{S(-kV, k)} \exp(-ikx'). \tag{18}$$

The integrand can be represented as:

$$\frac{Q(-kV, k)}{S(-kV, k)} = \frac{-Vi\eta\left(k - \frac{i}{V\eta}\left(\frac{V^2}{c_t^2} - 1\right)\right)}{S_1(V, k)},$$

where the function in the denominator

$$S_1(V, k) \equiv S(-kV, k)/k^2$$

is a cubic polynomial in k, the roots of which k_i can be determined using Cardano formulae. We will consider them known; Figure 1 shows the dependence of the roots k_i on the velocity V.

By taking the integral in (18) by residues, the expression for the function $f(V)$ in its final form is derived:

$$f(V) = -\sum_{\mathrm{Im}(k_i)>0} \left[\frac{1}{k} \frac{\left(k + \frac{i}{V\eta}\left(1 - \frac{V^2}{c_t^2}\right)\right)(k - k_i)}{(k - k_1)(k - k_2)(k - k_3)}\right]\Bigg|_{k=k_i}. \tag{19}$$

It should be noted that the dependence of the function $f(V)$ on velocity is continuous (see Figure 2, *a*)) despite the fact that at certain values a change in the roots that contribute to (19) takes place.

Let us now consider a typical experimental mode with the isotherm velocity of $V_{iso}(t) = \mathrm{const} \equiv V_p$. In this case, the equation of IPB motion (5) in the accompanying coordinate system, taking into account (13) and (16), takes the following form:

$$\frac{1}{2} - f(V) + \sum_{n=1}^{\infty} M_n \partial_t^n V - a\left\{\nabla T\left[X(t) - V_p t + h_o\right]\right\} = 0. \tag{20}$$

Equation (20) has an obvious solution with a constant IPB velocity equal to the isotherm velocity, $V = V_p$, as well as with a time-independent „lag“ of the IPB position $h_o < 0$ from the isotherm coordinate $V_p t$, equal to

$$h_o = \frac{1}{a\nabla T}\left[\frac{1}{2} - f(V_p)\right]. \tag{21}$$

In the general case, when the experimentally specified velocity of the isotherm is not constant, $V_{iso} = V_{iso}(t)$, the equation of IPB motion can be written in the following form:

$$\frac{1}{2} - f(V) + \sum_{n=1}^{\infty} M_n \partial_t^n V$$

$$- a\left\{T\left[X(t) - X_{iso}(t) + h_o(t)\right]\right\} = 0. \tag{22}$$

Analysis of the nature of possible solutions to this nonlinear differential equation of infinite order is facilitated by the fact that the dimensionless temperature gradient included in it is very small. Indeed, in dimensional quantities it is equal in order of magnitude to $\frac{b}{T_c}\nabla T_{\exp} \sim 10^{-8}$, where b is length of the lattice constant, and $\nabla T_{\exp}$ is temperature gradient specified in the experiment.

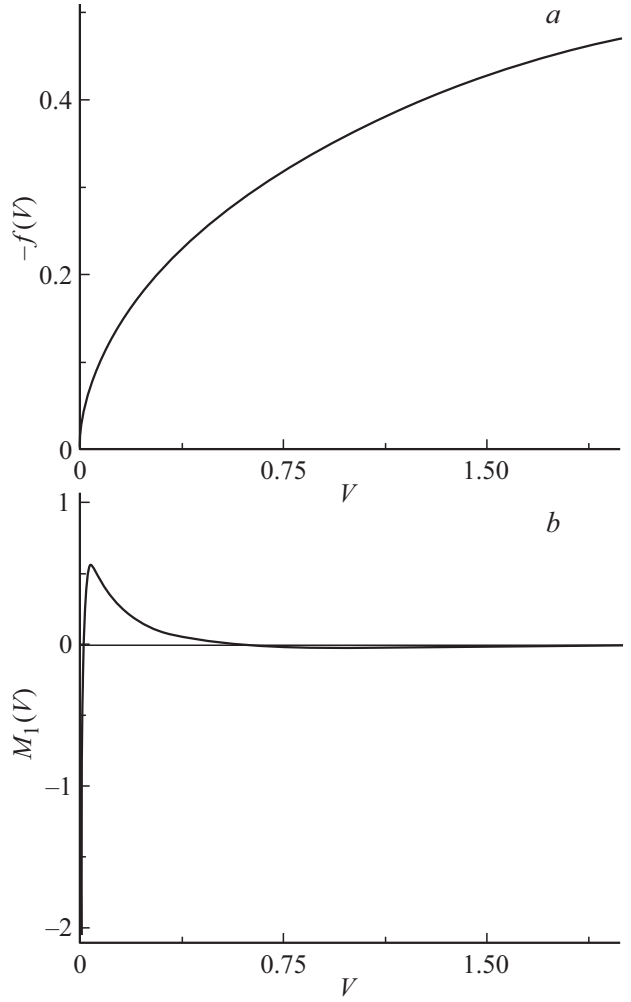

Figure 2. Dependence *a*) of the „friction force" — $f(V)$ by formula (19) and *b*) of the „mass" $M_1 = -C_1(V)$ by formula (24) on velocity for the following parameters: $\eta = 100$, $d = 10$, $c_t = 1400$.

When changing over to more convenient dimensionless variables of length and time, $x\nabla T \to x', t\nabla T \to t'$, the $f(V)$ function in (22) will not change (because the velocity in the new variables will remain the same), and „hypermasses" of the n-th order M_n will be preceded with small dimensionless factors $(\nabla T)^n$. Therefore, it is natural to limit the resulting asymptotic series taking into account only the term with $n = 1$.

As a result, (22) turns into a nonlinear differential equation of the 2-nd order, in which $M_1 = M_1(V)$. This allows, in particular, analyzing the linear stability of solutions to this equation in a standard way, i. e. studying the dependence of small deviations from them $h(t)$ at long times.

For example, in the case of a constant velocity of isotherm, $V_{iso}(t) = V_p$, the linearized equation is that of an oscillator with friction, with the coefficient of friction equal to the derivative $\left(\frac{df}{dV}\right) \equiv F'$ taken at $V = V_p$:

$$\nabla T M_1(V_p)\ddot{h} - F'(V_p)\dot{h} - ah = 0, \qquad (23)$$

where the dependence $M_1(V_p) < 0$ is determined through the corresponding coefficient $C_1(V)$ in (14):

$$M_1(V) = \frac{1}{4\pi i}\frac{\partial^2}{\partial V^2} \times \left[\int_{-\infty}^{\infty} dk \int_{-\infty}^{0} dx' \frac{Q(-kV, k)}{kS_1(V, k)} \exp(-ikx')\right]. \qquad (24)$$

As noted above, $a < 0$. As for the signs of $M_1(V_p)$ and $F'(V_p)$, for the values of the model parameters studied, the sign of the oscillator „mass" in (23), equal to $-C_1(V)$, is positive, as well as the sign of the „friction coefficient", $-F'(V_p)$, so that the deviations $h(t)$ decay exponentially over large times. However, it should be noted that with some other sets of model parameters, the „friction coefficient" may, in principle, turn out to be negative. In the low velocities extreme $V_p \to 0$, this, however, is impossible due to the fundamental requirement that entropy production should be positive (for more details on this issue, see the „Discussion" section in [2]).

Like the $f(V)$ function in (19), the $M_1(V)$ function can also be written in general form through the $k_(i)$ roots of the $S_1(V, k)$ function:

$$M_1(V) = \frac{i}{2}\frac{\partial^2}{\partial V^2} \times \left[\sum_{\mathrm{Im}\,(k_i)>0} \frac{\left(k - \frac{i}{V\eta}\left(\frac{V^2}{c_t^2} - 1\right)\right)(k - k_i)}{k^2(k - k_1)(k - k_2)(k - k_3)}\right]\Big|_{k=k_i}. \qquad (25)$$

Formula (25) was derived from (24) as a result of shifting the pole $k = 0$ into the lower half-plane by $i\epsilon$ $(\epsilon > 0)$ to regularize the expression in square brackets. Then the integral over k was calculated by residues, after which the remaining trivial integral over x' was taken. Figure 2 *b*) shows the dependence of mass $M_1(V)$.

Since substituting the roots $k_(i)$ using the Cardano formula results in a very cumbersome expression for $M_1(V)$, we do not write out the corresponding formula here.

It can be seen from the calculations of $M_n(V, d, \eta)$ in the general case, that these values quickly decrease with increasing velocity V and number n. It follows herefrom that the quantitative results obtained using the approximation of $M_1(V)$ in (22) improve with increasing IPB velocity, i. e. with increasing sample cooling rate.

If the sample is cooled at a time-dependent rate $\dot{T} = \dot{T}(t)$, then it is convenient to write the isotherm velocity in the form of $V_{iso}(t) = V_p + v(t)$, distinguishing its constant component V_p. Then the equation of IPB motion (22) in the approximation of $M_1(V)$ becomes an equation for an oscillator with mass and friction force depending on the velocity $V = V_p + \dot{h}$, which is subject to the variable

external force $R(t)$:

$$\nabla T M_1(V_p + \dot{h})\ddot{h} - f(V_p + \dot{h}) + f(V_p) - ah = a\int_0^t v(\tau)d\tau \equiv R(t). \quad (26)$$

It should be noted that for many ferroelectrics, a jump in spontaneous polarization at the PT point is accompanied by its weak increase with a further decrease in temperature [22]. In our model, this corresponds to the smallness of the coefficient $(-a)$. For such materials, equation (26) should work quantitatively especially well, because in this case, the dimensionless small parameter in expansion (22) becomes the product $(-a\nabla T)$.

5. Conclusion

This study analyzes the dynamics of an interphase boundary (IPB) entrained by an isotherm moving at a given velocity $V_{iso}(t)$ in a ferroelectric material having a piezoelectric effect in the paraphase. Within the framework of a model where the dependence of thermodynamic potential on polarization is given by two intersecting parabolas, we derived an approximate nonlinear differential equation of the 2-nd order for the IPB position coordinate $X(t)$. Our estimates have shown that this equation is applicable to describe the dynamics of the IPB for almost all actually used modes of isotherm movement $V_{iso}(t)$. In the mode usually used in experiments with a constant velocity of the isotherm, the movement of the IPB accompanying it at the same velocity turns out to be stable. It should be noted that the IPB dynamics can change qualitatively if the crystal contains a large number of defects that create an additional force of „dry friction“, pinning the boundary [31]. The correspondence of the parameter values of the 2-parabola model used in our calculations with the parameters used in the standard Landau model for PTs of the 1-st kind is indicated. This makes it possible to use experimentally known parameter values for a specific material to calculate the dynamics of the IPB within the equation (22) derived in the study.

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Conflict of interest

The authors declare that they have no conflict of interest.

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