ON THE CALCULATION OF DOMAIN WALL MOTION IN PEROVSKITE-TYPE FERROELECTRIC CRYSTALS

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The domain wall motion in perovskite-type crystals is investigated using nonlinear thermodynamics of irreversible processes. Equations of the velocity of motion of the domain wall on the applied electric field both for phase transitions of the first and second order are obtained. The obtained equations differ qualitatively for phase transitions of first and second order. For the first order phase transition, there is a linear dependence of the velocity of the 180° domain wall on the external electric field, but the temperature merely changes the coefficient of linear proportionality without affecting the nature of the functional dependence. The calculations are in full agreement with available experimental data.

1. Introduction

Switching polarization of a ferroelectric crystal, due to an electric field, is not homogeneous, but is affected by the generation of domains and their growth thanks to the movement of domain walls. The model theories for this phenomenon are based on a series of arbitrary assumptions concerning the dependence of growth on time, field, temperature, appropriate mechanical properties of the walls etc. However, the given problem can be solved only in terms of nonlinear thermodynamics of irreversible processes.

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2. Formulation

The Gibbs free energy per unit volume of a ferroelectric crystal may be represented in the Landau form

$$\Phi = \Phi_0 + \tfrac{1}{2}\alpha'_0(T-T_k)P^2 + \tfrac{1}{2}\beta P^4 + \tfrac{1}{3}\gamma P^6 + \tfrac{1}{2}k(\nabla P)^2 - EP, \tag{1}$$

where α'_0 is related to the Curie-Weiss constant, β and γ are Landau expansion coefficients, k describes the energy due to a nonhomogeneous order parameter P (polarization), T_k is the Curie-Weiss temperature and E stands for the external field. The temperature dependence of coefficients β, γ and k is neglected. The Gibbs free energy of the system is obtained as an integral over the volume V and represents a polarization function

$$\psi = \int \Phi(v)\,dv. \tag{2}$$

The evolution of the order parameter P towards its equilibrium value is described by the kinetic equation

$$\frac{\partial P}{\partial t} = -L\frac{\delta\psi}{\delta P} = -L\left[\frac{\partial\Phi}{\partial P} - \nabla\frac{\partial\Phi}{\partial(\nabla P)}\right], \tag{3}$$

where it is assumed that the kinetic coefficient L has no singularities near the phase transition point.

In the case of first order phase transition ($\beta < 0$) it is convenient to perform the following substitutions

$$\eta = \beta(\gamma k)^{-1/2}r \quad \tau = L\beta^2\gamma^{-1}t \quad P^2 = -\beta\gamma^{-1}s^2$$

$$E = -\beta^{5/2}\gamma^{-3/2}e \quad \theta = (-\beta)^{-2}\alpha'_0\gamma(T-T_k),$$

which permits one to reduce Eq. (1) to

$$\varphi(s) = \tfrac{1}{2}\theta s^2 - \tfrac{1}{2}s^4 + \tfrac{1}{6}s^6 - es, \tag{4}$$

where η, τ, s, e and θ are the reduced radius-vector, time, order parameter, electric field and temperature, respectively. At the phase transition point T_0, $\theta = 0.75$ because the relation between the thermodynamic expansion coefficients at this point is

$$3\beta^2 = 4\alpha'_0(T_0 - T_k)\gamma.$$

The general formula for θ is

$$\theta = \tfrac{3}{4}(T-T_k)(T_0-T_k)^{-1}. \tag{5}$$

For second order phase transition ($\beta > 0$) one can neglect the term proportional to the sixth power of polarization and performing the following substitutions

$$\eta = k^{1/2}r, \quad \tau = Lt, \quad P^2 = s^2(2\beta)^{-1}, \quad E = (2\beta)^{-1/2}e, \quad \theta = \alpha'_0(T-T_0),$$

we can rewrite Eq. (1) as:

$$\varphi(s) = \tfrac{1}{2}\theta s^2 + \tfrac{1}{4}s^4 - es \tag{6}$$

Equation (3) may be reduced to

$$\frac{\partial s}{\partial \tau} = \Delta_\eta s - \frac{\partial \varphi(s)}{\partial s},$$ (7)

where $\varphi(s)$ is given by Eq. (4) or (6). Equation (7) presents a nonlinear diffusion equation on the basis of which the main laws ruling the domain wall movement may be studied.

3. Results

By studying stationary crystal states by means of Eq. (7) it is possible to provide for each value of θ such a value $e = e_m$ that for $e > e_m$ Eq. (7) will have a single solution corresponding to a homogeneous polarization of the crystal. For the values $e < e_m$ Eq. (7) has two solutions s_1 and s_2. In this region of values the solution of Eq. (7) describing the motion of 180° domain wall, is a function $s(\tau, \eta_1)$ with the asymptotics

$$s_1(\tau, -\infty) = s_1, \quad s_2(\tau, +\infty) = s_2, \quad \frac{\partial s(\tau, \eta_1)}{\partial \eta_1}\bigg|_{\eta_1 = \pm\infty} = 0,$$ (8)

where η_1 is the projection of η on the axis perpendicular to the frontal part of the wall. The solution of a nonlinear equation of the type of Eq. (7) with boundary conditions (8) describes a front, stationary within the moving coordinate system $\eta^* = \eta_1 + u\tau$, where u is the velocity of the front, being in this case a function of θ and e. In other words the function $s(\tau, \eta_1)$ can be represented as $s(\tau, \eta_1) \equiv s(\eta^*)$ what describes the stationary state of domain wall, which is mooving with speed u along the axis η_1. To be specific, let us presume that $e \geqslant 0$, $s_2 \geqslant -s_1$. In the moving coordinate system Eq. (7) reads into

$$\frac{d^2 s}{d\eta^{*2}} = u\frac{ds}{d\eta^*} + \frac{\partial \varphi(s)}{\partial s}$$ (9)

with boundary conditions

$$s(-\infty) = s_1, \quad s(+\infty) = s_2, \quad \frac{ds}{d\eta^*}\bigg|_{\eta^* = \pm\infty} = 0.$$ (10)

Solution $s(\eta^*)$ gives the possibility of finding the velocity of the domain wall motion. From Eqs. (9) and (10) follows

$$u\int_{-\infty}^{\infty} \left(\frac{ds}{d\eta^*}\right)^2 d\eta^* = -\int_{-\infty}^{\infty} \frac{\partial \varphi}{\partial s}\frac{\partial s}{\partial \eta^*} d\eta^* = \varphi(s_1) - \varphi(s_2).$$ (11)

On the other hand, the function $s(\eta^*)$ determined from Eq. (9) depends parametrically on u. This can be used for an approximate determination of u by the variational method. In the approach using the local potential, Eq. (9) is obtained from the condition of integral

stationarity

$$I = \int_{-\infty}^{\infty} L\,d\eta^*, \tag{12}$$

where

$$L = L_1 + us\frac{ds^0}{d\eta^*}, \quad L_1 = \frac{1}{2}\left(\frac{ds}{d\eta^*}\right)^2 + \varphi(s). \tag{13}$$

The index zero in Eq. (13) designates the function which is held constant. We choose for a test function

$$s(\eta^*) = \begin{cases} s_1 & \eta^* \leqslant \eta' \\ \eta^*/z & \eta^* \in (\eta', \eta''), \end{cases} \tag{14}$$
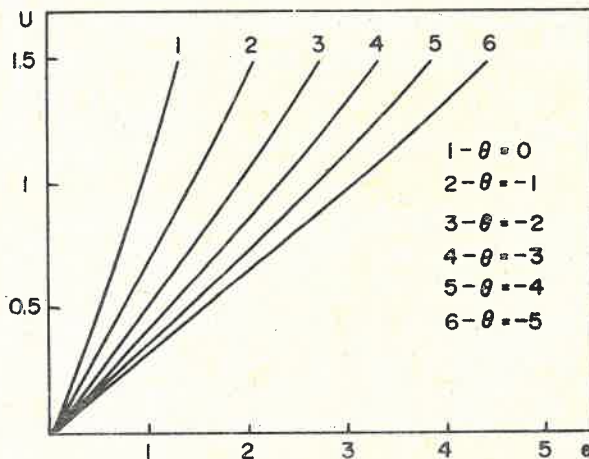

Fig. 1. Dependence of the velocity of motion of the domain wall on the applied electric field at varying temperature in the case of first order phase transitions
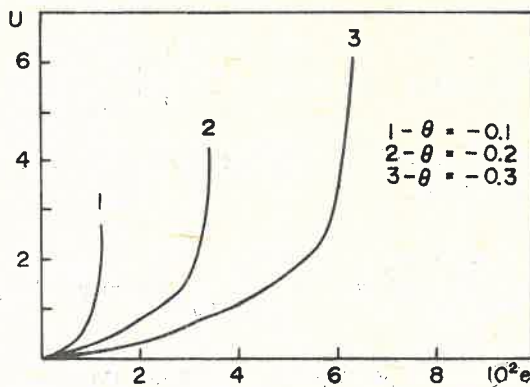

Fig. 2. Dependence of the velocity of motion of the domain wall on the applied electric field at varying temperature in the case of second order phase transition

where $\eta' = zs_1$, $\eta'' = zs_2$ but z is the variational parameter derived from the condition

$$\frac{dI}{dz} = 0 \qquad (15)$$

By substituting Eq. (14) into Eq. (11) and with the help of condition (15) we obtain nonlinear equations for the velocity of the domain wall from where one has

$$u = \frac{\varphi(s_1) - \varphi(s_2)}{(s_2 - s_1)^{1/2}\{(s_1 + s_2)\left[\varphi(s_2) - \varphi(s_1)\right] + 2[\int\limits_{s_1}^{s_2} \varphi(s)ds - \varphi(s_2)s_2 + \varphi(s_1)s_1]\}^{1/2}}. \qquad (16)$$

For $e > 0$, we get $u > 0$ which corresponds to the spreading of domains with the polarization s_2. The results calculated by formula (16) are produced in Figs. 1 and 2 for phase transitions of the first and second order, respectively.

4. Discussion

As is seen from Figs. 1 and 2 the dependence of the velocity of the motion of the domain wall differs qualitatively for phase transitions of first and second order. In the case of first order phase transition, there is a linear dependence of the velocity of the 180° domain wall on the external electric field, but the temperature, at which the crystal is analysed, merely changes the coefficient of linear proportionality without affecting the nature of the functional interrelation. It must be emphasized that the obtained results of calculation are in full agreement with the available experimental data [1]. Direct comparison shows that the experimental curves are shifted to the right and down with respect to the theoretical curves. Hence, when the theoretical dependence has the form $u = u(E)$, the experimental one will be $u = u(E-E_0)$, which can be accounted for by the presence of defects in the crystal, the latter form traps, the breaking from which requires some additional field E_0. Comparison of the calculated curves for second order phase transition with experimental data [2] also shows a full qualitative agreement. It should be pointed out that the experimental data imply that both for the first and second order phase transition, the velocity of domain walls considerably increases when the phase transition temperature is approached. The results of our calculations also show this velocity becomes a nonlinear function of temperature near the phase transition.

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