Smearing of phase transition due to a surface effect or a bulk inhomogeneity in ferroelectric nanostructures

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The boundary conditions, customarily used in the Landau-type approach to ferroelectric thin films and nanostructures, have to be modified to take into account that a surface of a ferroelectric (FE) is a defect of the “field” type. The surface (interface) field is coupled to a normal component of polarization and, as a result, the second order phase transitions are generally suppressed and anomalies in response are washed out. In FE films with a compositional (grading) or some other type of inhomogeneity, the transition into a monodomain state is suppressed, but a transition with formation of a domain structure may occur.

Theoretical studies of phase transitions in thin films and the corresponding size effects within the Landau theory \([1,2]\) have been undertaken since 1950s. Recently the interest to these questions has risen dramatically in view of the applications of ferroelectric thin films \([3]\) and a discovery of various ferroelectric nanostructures \([4]\). The boundary conditions for thin films were originally discussed by Ginzburg and Landau (GL) in 1950 \([5]\) and by Ginzburg and Pitaevskii in 1958 \([6]\). It was shown by GL that, if the properties of the boundary layer are the same as of the bulk, one arrives at the condition that the gradient of the order parameter vanishes at the surface, \(\nabla_\perp \eta = 0\) (in zero magnetic field, \(\vec{n}\) is the normal to the surface). Starting from a microscopic theory, de Gennes has shown that for a superconductor-metal interface with no current and magnetic field a more general boundary condition applies, \(\nabla_\parallel \eta + \eta/\delta = 0\), where \(\delta\) is the characteristic length scale describing the proximity effect \([7]\). These conditions are very general and were obtained phenomenologically by Kaganov and Omelyanchouk for a surface of a ferromagnet \([8]\) (cf. review in \([9]\)). Kretschmer and Binder \([10]\), using the same boundary conditions, have taken into account the depolarizing field, which is important when a ferroelectric polarization (or magnetization) is perpendicular to the surface. Later these boundary conditions have been used customarily in studies of phase transitions in ferroelectric films (see, e.g. \([11]\)).

It is obvious, however, that while the treatment \([8,10]\) is appropriate for magnetics, it overlooks an important specific feature of ferroelectric phase transitions in thin films, wires, and other systems with boundaries. Indeed, there is an effective field at the surface (interface) of any material appearing because the surface breaks the symmetry of the bulk. For instance, a part of this surface field might be due to a Coulomb dipole field (double layer), contributing to the work function \([12,13]\). This makes ferroelectric surfaces qualitatively different from the surfaces of magnetics. The effective field is coupled to the component of polarization perpendicular to the surface/interface and, as a result, the second order ferroelectric phase transitions are typically smeared out, as we shall see below.

We shall discuss, as an example, a paraelectric-ferroelectric phase transition in cubic perovskite thin films where a surface is perpendicular to one of the cubic axes. The surface or interface eliminates all the symmetry elements, which change a vector perpendicular to the surface and generates a local field conjugated to the polarization component perpendicular to the surface (\(P_z\) in our case) \([14]\). To demonstrate the effect, we consider the state with \(P_z\), \(P_y = 0\), described for given potentials on electrodes \([15,16]\) by the free energy \(\tilde{F}_f = F_{LGD} + \int dV E_\parallel^2/8\pi - \sum_\alpha e_\alpha \varphi_\alpha\),

\[
F_{LGD} = \int dV \left[ \frac{1}{2} AP^2 + \frac{1}{4} BP^4 + \frac{1}{2} g \left( \frac{dP}{dz} \right)^2 + \frac{1}{2} D (\nabla_\perp P)^2 \right],
\]

(1)

where \(\nabla_\perp = (\partial/\partial x, \partial/\partial y)\) is the gradient in a plane of the film, \(q_a (\varphi_\alpha)\) are the charges (electrostatic potentials) at the electrodes \(a = 1, 2\). Here \(\nabla_\perp = 0\) for the monodomain state. We assume ideal electrodes with a vanishing Thomas-Fermi screening length. As discussed above, the surface produces an effective surface field \(w\), and, generalizing Ref. \([8]\), we have to add the surface energy to (1) to obtain the free energy of the film

\[
\tilde{F} = \tilde{F}_f + \int dS \left( \frac{1}{2} \alpha P^2 - wP \right),
\]

(2)

where \(\alpha\) corresponds to a “temperature”-like component of the surface energy. We obtain from Eqs. (1) and (2) after an integration by parts the correct boundary conditions for ferroelectrics

\[
\alpha_{1(2)} P + (-) g \frac{dP}{dz} = w_{1(2)}, \quad z = \pm (l/2).
\]

(3)
One can estimate that $\alpha \sim d_{\text{at}}$, where $d_{\text{at}}$ is the characteristic “atomic” length scale, on the order of the lattice constant. The electric field at the surface, first considered many decades ago [12], is on the order of $w/d_{\text{at}} \sim \Phi_s/d_{\text{at}} \sim 1V/\AA \approx 10^8V/cm$, where $q\Phi_s \sim 4eV$ is the typical workfunction for ferroelectrics [3]. The surface bias field corresponds to a surface charge $\sim 100\mu C/cm^2$, which is on the order of an “atomic” polarization $P_{\text{at}} = q/d_{\text{at}}^2 \sim 200\mu C/cm^2$, so that $w \sim P_{\text{at}}d_{\text{at}}$ (we expect that the non-Coulomb contribution to $w$ is of the same order of magnitude). The polarization $P (z)$ is found from the equation of state for (1) and the Poisson equation, assuming that there is no external charge, and neglecting for a moment the non-linear terms in polarization:

$$AP - g \frac{d^2P}{dz^2} = E, \quad (4)$$

$$\frac{d}{dz} (E + 4\pi P) = 0, \quad (5)$$

$$\frac{1}{l} \int_1^2 Edz = \frac{\varphi_1 - \varphi_2}{l} = U/l \equiv E_0, \quad (6)$$

where $E_0$ is the external electric field. We obtain from Eqs. (5) and (6)

$$E = E_0 - 4\pi [P(z) - \bar{P}], \quad (7)$$

where the overbar means an average over the film, i.e. $\bar{f} = (1/l) \int_{l/2}^{l/2} dzf(z)$. Substituting this into Eq. (4) and integrating over the film, we find

$$\lambda \frac{\bar{P}}{2} - g \int_0^2 \frac{(dP(l/2)}{dz} - \frac{dP(-l/2)}{dz} = E_0. \quad (8)$$

We write down the solution as a sum $P = P_0 + p(z)$ of the homogeneous, $P_0 = (E_0 + 4\pi \bar{P})/(A + 4\pi)$, and the inhomogeneous, $p(z)$, term

$$p = C_1 \exp[-\lambda(z + l/2)] + C_2 \exp[-\lambda(l/2 - z)], \quad (9)$$

$$C_{1(2)} = (w_{1(2)} - w_{1(2)} P_0)/(\alpha_{1(2)} + \lambda g), \quad (10)$$

where $\lambda = \sqrt{A + \pi g} \approx \sqrt{\frac{4\pi g}{l} \sim d_{\text{at}}^{-1}}$. Since $P = P_0 + (C_1 + C_2)/\lambda l$, we obtain with the use of Eqs. (10),(8)

$$A' \bar{P} = E' + \frac{\lambda g}{l} \left( \frac{w_1}{\alpha_1 + \lambda g} + \frac{w_2}{\alpha_2 + \lambda g} \right), \quad (11)$$

where $A' = A(1 - \xi/\lambda l) + \lambda g \xi/l \approx A + 4\pi \xi/\lambda l$, $E' = E_0(1 - \xi/\lambda l)$, and $\xi = \alpha_1/(\alpha_1 + \lambda g) + \alpha_2/(\alpha_2 + \lambda g) \sim 1$. The phase transition in this case is smeared out, since generally the surface dipoles are asymmetric. In the symmetric case, $w_1 = -w_2$, $\alpha_1 = \alpha_2$, the phase transition persists, but the transition temperature of a transition into a monodomain state is shifted by the amount inversely proportional to the thickness of the film, with the following estimate for displacive systems (cf. Ref. [10]):

$$\Delta T_c = \frac{4\pi \xi T_{\text{at}}}{\lambda l}. \quad (12)$$

The monodomain transition in the symmetric case occurs at $A = -\lambda g \xi/l \approx -d_{\text{at}}/l$. This is close to a transition with the formation of domains [17]. Which transition actually occurs depends on materials parameters.

The surface dipoles discussed above are a special case of polarization due to gradients of a scalar quantity (concentration $c$, density, temperature, etc.) and they are accounted for by a term like

$$f_c = -\gamma \frac{\partial \Phi}{\partial c}, \quad (13)$$

in the free energy, where the coefficient $\gamma$ is estimated as $\gamma \sim P_{\text{at}}d_{\text{at}}$ [18,19] (see also [20]).

Consider now the case of a film with a compositional profile (grading) given by e.g. the concentration of one of the components of a ferroelectric alloy $c = c(z)$. Such systems are currently a focus of research in ferroelectrics due to their unusual pyroelectric characteristics [21]. The equation of state of the graded ferroelectric film is

$$A(z)P + BP^3 - g \frac{d^2P}{dz^2} - D\nabla^2 P = E_0 + 4\pi (\bar{P} - P) + \gamma \frac{dc}{dz}. \quad (14)$$

Consider a special case of a step-wise concentration profile, i.e. $c = c_1$ when $0 < z < l_1$, and $c = c_2$ when $-l_2 < z < 0$, and the boundary conditions are “neutral” ($dP/dz = 0$ at $z = l_1, -l_2$). The equation of state in this case is

$$A_r P + BP^3 - g \frac{d^2P}{dz^2} = E_0 + 4\pi (\bar{P} - P), \quad (15)$$

for the both parts of the film $r = 1, 2$. The boundary conditions at $z = 0$ follow from the continuity of a displacement field $E + 4\pi P$ and the equation of state (14). In displacive systems the electric field $E \sim AP$ is always much smaller than the polarization $P$, since $|A| \ll 1$ [17]. Hence, with high accuracy $|c_1 - c_2|/4\pi \ll 1$, the boundary conditions are

$$g \left( \frac{dP_1}{dz} - \frac{dP_2}{dz} \right) = -h, \quad (16)$$

at $z = 0$, with $h = \gamma (c_1 - c_2) \approx P_{\text{at}}d_{\text{at}}(c_1 - c_2)$.

We have studied before [17] a similar situation but without the concentration gradient, i.e. for $\gamma = 0$. Having assumed that the $z$-dependence of $A$ is due to the concentration dependence of the Curie temperature with $T_{\text{Curie}} \sim T_{\text{Curie}}(c_1 - c_2)$ and $dA/dT \sim T_{\text{Curie}}^{-1}$ (displacive systems) we have shown that for $(c_1 - c_2) \gtrsim d_{\text{at}}/l$ the loss of stability of a paraphase occurs with a formation of a domain structure, and it takes place at $A_2 > 0$ but $A_1 < 0$ and $|A_1| \sim (c_1 - c_2) \gtrsim d_{\text{at}}/l$ [17]. For these
threshold values of concentration inhomogeneity the result of Ref. [17] stands, whereas in the case of the bulk inhomogeneity and general nonsymmetric boundary conditions the results are different (see below).

The main effect of the bias field is that now there is a polarization at all temperatures, and, therefore, the phase transition into a monodomain state is smeared out. However, a phase transition with formation of a domain structure is still possible. To see this, we need to investigate a stability of a monodomain solution of (14). First, we need to find the average polarization \( \bar{P} \) across the film. Integrating Eq. (15) over the film thickness, we obtain

\[
\bar{A}P + B\bar{P}^3 + \delta A_\nu \delta P + B \left( 3\bar{P}\delta P^2 + \delta \bar{P}^3 \right)
\]

\[
= E_0 + w/l,
\]

\[
w = h + w_1 + w_2 - \alpha_1 P(l_1) - \alpha_2 P(-l_2),
\]

where \( \bar{A} = \nu_1 A_1 + \nu_2 A_2 \), \( \delta A_\nu = A_\nu - \bar{A}, \nu_1(2) = l_1(2)/l, \)

\( l_1 = l_1 + l_2, \) Fig. 1. There are two possibilities: (i) near symmetric, \( |w_1 + w_2| \lesssim |h| \) and (ii) asymmetric surfaces, \( |w_1 + w_2| \gg |h| \). In the first case the monodomain transition is smeared out by the presence of the gradient dipole field \( h \), but the transition with domain formation is possible. In the second, more general, case the monodomain transition is smeared out, and domains either form or they do not, depending on the concentration gradient and/or the thickness of the film.

Importantly, the effective bias field \( w/l \), conjugated to the average order parameter \( \bar{P} \), Eq. (17), is large. For a 1000Å thick film it would have the same effect as if there were an external electric field \( \sim 10^5 \) V/Å, which, for comparison, is only marginally smaller than the breakdown field in graded FE \( E_b \approx 0.75 \cdot 10^6 \) V/cm [21].

To investigate a stability loss of a parahase, we assume that the linear approximation is valid and check later if the solution justifies the assumption. We estimate \( \bar{P} \approx w/Al \sim P_{at}d_{at}/(c_1 - c_2)l \), hence the first term in (17) is \( \bar{A}P \sim w/l \sim P_{at}d_{at}/l \). The cubic term is \( BP_3^3 \sim P_{at}^{-2} (w/Al)^3 \sim P_{at} (d_{at}/Al)^3 \), since \( w \sim P_{at}d_{at} \), and we can neglect it in comparison with the first linear term in (17) when \( \bar{A} \sim (c_1 - c_2) \gg (d_{at}/l)^{2/3} \). The latter is the condition for the linearization of the equation of state (14), which takes the form

\[
\left( A_r + 4\pi \right) \delta P - gd^2 \delta P/\delta z = -w/l - \delta A_r \bar{P},
\]

with a solution

\[
(p_1(z) - w/l + \delta A_r \bar{P})/A_r + 4\pi = p_1(z),
\]

\[
p_1 = ae^{-\lambda_1 z} + C_1 e^{-\lambda_1(l_1 - z)}, \quad 0 < z < l_1,
\]

\[
p_2 = be^{\lambda_2 z} + C_2 e^{-\lambda_2(l_2 - z)}, \quad -l_2 < z < 0.
\]

We can replace \( \lambda_1 = \lambda_2 = \lambda = \sqrt{4\pi/g} \) and obtain from the boundary conditions \( a = b = h/(2\lambda g) \) and

\[
\lambda g C_1 + \alpha_1 P(l_1) = w_1,
\]

\[
\lambda g C_2 + \alpha_2 P(-l_2) = w_2.
\]

A reasonable approximation is \( P(l_1) \approx C_1 = w_1/\alpha_1 + \lambda g \), \( P(-l_2) \approx C_2 = w_2/\alpha_2 + \lambda g \), Fig. 1. From first-principles calculations at the surfaces of BaTiO_3 and PbTiO_3 \( P \sim 10^{-1} P_{at} \) (see, e.g. [22]).

Consider the third and fourth terms on the left hand side of Eq. (17). With the use of Eqs. (20)-(22) we obtain the estimate \( \delta A_\nu \bar{P} \sim (c_1 - c_2)w/4\pi l \ll \bar{A} \bar{P} \), since there is an additional small factor \( (c_1 - c_2)/4\pi \). Both terms in \( \delta P_r(z) \) give contributions to this estimate of the same order of magnitude. The term \( BP_3^3 \bar{P}^2 \sim P_{at}^{-2} \delta P^3 \) is the condition that it is smaller than \( \bar{A} \bar{P} \) reads \( \bar{A} \sim (c_1 - c_2) \gg (d_{at}/4\pi l)^{3/2} \), and it is certainly obeyed when \( (c_1 - c_2) \gg (d_{at}/4\pi l)^{3/2} \), which is the condition for the linearization, obtained above. The last term in (17) is very small if \( (d_{at}/4\pi l)^{3/2} \ll 1 \), which is always the case.

Now, we shall see if the domain formation is possible. Following the procedure of Ref. [17], we have to linearize (14) about the monodomain solution (inhomogeneous along z direction only) and look for its non-trivial solutions in the “soft” part of the film with \( A_1 < 0 \) in the form of the “polarization wave”, \( P(x, z) = \bar{P} + \delta P(z) + \xi(x, z) \), where \( \xi(x, z) \propto e^{ikz} \). We arrive at the same problem as in Ref. [17] but with a renormalized coefficient \( A_1 \rightarrow A_1 = A_1 + 3B_0 P_2 \). The boundary conditions for \( \xi(x, z) \) are exactly the same as in Ref. [17], and \( A_1 \) enters the condition for instability \( (A_2 > 0 \) for the “hard” part does not). Domain formation is possible when \( A_1 \sim -d_{at}/l \), in spite of a positive renormalization. The condition for this reads \( A \gg (d_{at}/l)^{1/2} \). This condition is stricter than the one for the linearity of the equation of state (17), meaning that our using of the linearized equation for \( P \) is justified. There is also a range of concentration gradients, \( (d_{at}/l)^{2/3} \sim A \sim (c_1 - c_2) \sim (d_{at}/l)^{1/2} \), when one can
linearize the equation of state for $\tilde{P}$, but domains do not form ($A_1 > 0$). Finally, when the concentration gradient is even smaller, $A < (d_{at}/l)^{2/3}$, there is no domain formation and the equation for $\tilde{P}$ is substantially nonlinear, $\tilde{A}P < BP^3$. Therefore, the phase transition into a monodomain state occurs when, in general, the concentration gradient is large enough, $c_1 - c_2 > (d_{at}/l)^{1/3}$, or if, for a given concentration gradient, the film exceeds some critical thickness, $l > l_4 = d_{at}/(c_1 - c_2)^2$. If the system does split into domains in presence of the built-in surface bias field, the opposite domains will have different absolute values of polarization.

In a special case of symmetric surfaces, when $|w_1 + w_2| \lesssim |h|$, the domains always form. Here we find that the net polarization is due mainly to concentration inhomogeneity and is much smaller than in the general case considered above, $P \approx h/Al \sim P_{at}d_{at}/l$. This means that the term $BP^3$ is on the order of $P_{at}(d_{at}/l)^3$ (since $B \sim P_{at}^2$), at the same time the term $\tilde{A}P \sim (c_1 - c_2)P_{at}d_{at}/l$, i.e. for $\bar{A} \sim (c_1 - c_2) \gtrsim (d_{at}/l)^2$ the linear term indeed dominates in (17), $\tilde{A}P \gg BP^3$. We obtain also that $\delta P \approx h/(4\pi l) \approx \tilde{A}P/(4\pi) \approx (c_1 - c_2)\bar{P}/4\pi \ll P$. Because of this relation, all terms on the left hand side of Eq. (17) are indeed small in comparison with the first one, $\tilde{A}P$, and can be omitted. In the region of a stability loss with respect to domains $A_1 \sim -d_{at}/l$, therefore, $3BP^2 \sim (d_{at}/l)^2$, i.e. the positive renormalization of $A_1$ is very small, $\bar{A}_1 = A_1 + 3BP^2 \approx A_1 < 0$ and the system splits into domains. Therefore, for symmetric surfaces the presence or absence of the interfacial bias field at the boundary between two ferroelectric layers does not change our earlier prediction that practically any inhomogeneity, however small, would lead to a domain formation [17].

One should make a reservation in case the boundary conditions correspond to a “surface ferroelectricity” ($\alpha < 0$), then the monodomain transition can occur before a domain structure forms. However, this is a somewhat special case and, more importantly, effect of any real electrodes is rather opposite: it tends to suppress the ferroelectric transition into a monodomain state [23]. Therefore, it may be fairly difficult to observe the effects of a “surface ferroelectricity” in the case of spontaneous polarization normal to electrodes. Certainly, the real boundaries are never planar but rather rough. It seems likely that in real samples there are regions where the bias field is much smaller than would be in the case of planar boundaries. In these “weak” regions even small inhomogeneities in the materials constants would lead to a formation of domains, just as in the case of a sample with the “neutral” boundary conditions.

Note that even in a film with a step in a concentration profile the polarization is almost constant throughout the sample (with the exclusion of near-surface areas, Fig. 1). This is a result of a long-range depolarizing Coulomb field. It was neglected in a recent attempt to calculate the profile of polarization in the graded FE numerically, Ref. [24], and this led to erroneous conclusions. Therefore, those speculations do not apply to the observed behavior, like a large apparent pyroelectric coefficient. In particular, the build-in bias voltage due to polarization inhomogeneity, that has been calculated in [24], should be exactly zero, $4\pi \int dz \{P(z) - \bar{P}\} = 0$, Eq. (7). In fact, in graded samples there may be a bias voltage build-up due to a charge trapping, etc., and this could be related to the measured anomalous pyroelectric properties of these films, see e.g. a discussion in Ref. [25]. It is worth mentioning that dipoles, introduced by interfaces, are likely to be important in ferroelectric superstructures, where they can affect an electric response of the structures [26].

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