Effects of anisotropic elasticity in the problem of domain formation and stability of monodomain state in ferroelectric films

A.M. Bratkovsky$^1$ and A.P. Levanyuk$^{1,2,3}$

$^1$Hewlett-Packard Laboratories, 1501 Page Mill Road, Palo Alto, California 94304
$^2$Dept. Fiz. Mat. Cond., Universidad Autonoma de Madrid, Madrid 28049, Spain
$^3$Moscow Institute of Radioengineering, Electronics and Automation, Moscow 117454, Russia

(Dated: January 1, 2011)

We study cubic ferroelectrics films that become uniaxial with a polar axis perpendicular to the film because of a misfit strain due to a substrate. The main present result is the analytical account for the elastic anisotropy as well as the anisotropy of the electrostriction. They define, in particular, an orientation of the domain boundaries and stabilizing or destabilizing effect of inhomogeneous elastic strains on the single domain state. We apply the general results to perovskite systems like BaTiO$_3$/SrRuO$_3$/SrTiO$_3$ films and find that at least not far from the ferroelectric phase transition the equilibrium domain structure consists of the stripes along the cubic axes or at 45 degrees to them. We have also showed that in this system the inhomogeneous strains increase stability with regards to the small fluctuations of the metastable single domain state, which may exist not very close to the ferroelectric transition. The latter analytical result is in qualitative agreement with the numerical result by Pertsev and Kohlstedt [Phys. Rev. Lett. 98, 257603 (2007)], but we show that the effect is much smaller than those authors claim. We have found also that the electrostriction and anisotropic elasticity may decrease stability of a single domain state instead of increasing it. The single domain state is metastable at certain large thicknesses and becomes suitable for memory applications at even larger thicknesses when the lifetime of the metastable state becomes sufficiently large.

PACS numbers: 77.80.Dj, 77.80.bn, 77.55.Px

I. INTRODUCTION

Properties of domain structures in thin ferroelectric films is currently a focus of extensive research. It is expected, quite naturally, that an understanding and an ability to control these properties will determine the prospects of applications of nanometer-size ferroelectrics. It depends critically on the external conditions like presence or absence of electrodes. In this paper, we discuss domain structures in a system, which is, perhaps, the most important for applications: a ferroelectric film with electrodes. The polar axis of the material is perpendicular to the film plane and the electrodes are ‘real’ meaning that the electric field penetrates into them, although only over tiny depths < 1Å. This is an adequate model for the perovskite ferroelectric films on a substrate with compressive strain, like BaTiO$_3$/SrRuO$_3$/SrTiO$_3$ (BTO/SRO/STO)$^\dagger \ddagger$ where the misfit strain drives the FE film into a uniaxial state. We supplement our analytical results with the relevant numerical estimates for BaTiO$_3$ (BTO), PbTiO$_3$ (PTO), and Pb(Zr$_{0.5}$Ti$_{0.5}$)$_3$O$_3$ (PZT) using the material constants available in the literature.

Incompletely screening of the depolarizing field by SrRuO$_3$ electrode leads to an absolute instability of a single domain state and formation of a sinusoidal domain structure when thickness of BaTiO$_3$ film is slightly above the minimal thickness compatible with ferroelectricity in this system$^\dagger \ddagger$. It seems that this situation is typical of real electrodes and we shall consider this case only. To find the minimal thickness, one does not need to take into account higher order terms in the Landau-Ginzburg-Devonshire (LGD) free energy including the terms describing the electrostriction since the problem of stability of the paraelectric phase is linear$^\dagger \ddagger$. But in order to reveal the characteristics of this structure, i.e. to find out if the equilibrium structure is stripe-like or checkerboard and how the domain boundaries are oriented one has to take into account the anisotropy of elastic and electrostrictive properties of the ferroelectric. This is the main goal of the present paper. Specifically, we consider the case of cubic crystal anisotropy of elastic and electrostrictive properties only. This is relevant for films of cubic perovskites which become tetragonal because of in-plane misfit strains due to cubic substrates like in the above-mentioned system. The change of cubic anisotropy to tetragonal affects most strongly the dielectric properties since the crystals are “soft” dielectrically. They have much smaller effect on the elastic and electrostrictive properties, which can be considered to be the same as in cubic parent crystals.

Explicit account for the electrostriction and anisotropic elasticity is relevant also for study of stability of single domain state. This has been correctly pointed out by Pertsev and Kohlstedt$^{9,10}$ although these authors have missed several important points. Importantly, however$^\dagger \ddagger$, they overlooked that the state whose stability they were studying
was actually metastable. Therefore, its stability with respect to small fluctuations did not mean that this state can be used in memory applications. Indeed, its lifetime is very short if the film thickness is not sufficiently larger than that calculated by Pertsev and Kohlstedt as the limit of single domain stability. We shall also discuss this stability among other questions. This makes sense because of several reasons. First, Pertsev and Kohlstedt performed numerical calculations using material constants for BaTiO$_3$ and Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$ and the electrode parameters of SrRuO$_3$ while our results are analytical and apply to any material of the same symmetry. Moreover, our method applies to other symmetries as well. Second, Pertsev and Kohlstedt studied stability of the single domain state with respect to “polarization wave”-like fluctuations with a single specific direction of the wave vector, while we consider waves with k-vectors in arbitrary direction. Third, Pertsev and Kohlstedt apparently misinterpreted their own results by mixing together the well-known effect of homogeneous misfit strains and the effect of strains due to inhomogeneous polarization. In fact, the misfit strain simply results in renormalization of the materials constants and was effectively taken into account by all the previous authors. Only the account of the inhomogeneous polarization was pioneered in Ref.\textsuperscript{2}. We show that this effect was vastly overestimated by Pertsev and Kohlstedt. In fact, the formal difference by more than an order of magnitude between the results with and without account for electrostriction that they claimed stems from improper comparison of the compressed film with the materials constants renormalized by the misfit strain to one without any such renormalization at all, and not from the effect of the inhomogeneous strains on stability of single domain state.

Our analytical calculations provide a general view on the role of the inhomogeneous strains in stability of single domain state. In particular, they reveal a possibility which seems academic at the moment but no reason is seen to exclude it altogether. We mean a specific state where elasticity provokes domain formation of non-ferroelastic ferroelectric 180° domains. Such a state is realized if a certain condition on the electrostrictive and elastic constants is met. We are not aware of an experimental realization of these conditions but we cannot find arguments prohibiting them. It is worth mentioning that a qualitative conclusion about possibility of both stabilizing and destabilizing role of inhomogeneous elastic strains for single domain state in ferroelectric films on substrates has been made in our previous paper where we considered an academic case of a single electrostriction constant and assumed isotropic elasticity\textsuperscript{12}. A surprising result of the present work is that the destabilizing effect of the inhomogeneous strains may be very large contrary to the stabilizing one. Another unexpected result is the possibility of a checkerboard domain state if some conditions on the material constants are met. Let us mention that without account for the anisotropic polarization-strain coupling one comes to the conclusion about impossibility of such a state. For the perovskites this conclusion remains valid but not in the general case.

Studying the sinusoidal domain structure in BTO, PTO and PZT films on SrTiO$_3$, we find that the equilibrium orientation of the ”domain walls” is parallel (perpendicular) to the cubic axes in the film plane for BTO and PTO and is at 45° to these axes for PZT. In all cases the free energy of the sinusoidal domain structure depends very weakly on the domain wall orientation. This is mainly due to both systems being nearly isotropic elastically and, additionally, the relevant electrostriction constant is relatively small. This observation may be important for understanding domain creation at smallest thicknesses of the ferroelectric films. For BTO/SRO/STO system, our analytical calculations provide confirmation of the qualitative result of Pertsev and Kohlstedt about stabilizing effect of inhomogeneous elastic strains for single domain state in this system but with the above mentioned strong disagreement with their statement about importance of this effect.

Having mentioned advantages and new possibilities provided by analytical calculations we should mention also their inherent shortcomings. Our analytical method is feasible within a certain approximation only. This approximation implies that the domain period is less than the film thickness. This condition is fulfilled for thick enough films but in very thin films the two quantities are in fact comparable. Therefore, the accuracy of our calculations should be investigated for these films, so the new numerical studies are desirable. We do not expect, however, that the difference between the results of approximated and more exact calculations either within a continuous medium theory or within microscopic theories will be very large given close results of continuous and first principles theories even for films that are just several unit cell thick (see, e.g.\textsuperscript{2}).

The paper is organized as follows: we describe the approximations used and define the terms in the LGD free energy that can be neglected within our approximation in Sec\textsuperscript{III}. This let us avoid unnecessary lengthy formulas in the rest of the paper. We spell out the constituent equations in Sec\textsuperscript{III} and then solve the general problem for the ‘polarization wave’ (embryonic stripe domains) in the FE film with full account for elastic coupling. This is further used in Sec\textsuperscript{IV} to determine that the domain walls align with crystallographic cubic axes. Then, we find the conditions when the monodomain state first loses its stability with regards to the stripe domain structure in Sec\textsuperscript{VI}. One previously unexplored possibility is that the system can lose stability with regards to checkerboard domain structure, but our results in Sec\textsuperscript{VII} show that such a structure is absolutely unstable in perovskites although it is not necessarily so in the general case. We summarize the present results in the Conclusions.
II. OUTLINE OF THE METHOD AND THE APPROXIMATIONS USED

The main conclusions of this paper are made by analyzing the formula for free energy of the total system as a function of the amplitude $a$ of the ferroelectric "polarization wave" presenting the sinusoidal domain structure and the homogeneous part of the ferroelectric polarization, $p$. For the electrode and the film parameters of a system like BTO/SRO/STO, the ferroelectric polarization that is perpendicular to the film plane, schematic of which is shown in Fig. 1, has the form

$$P_z(x, y, z) = p + a \cos kr \cos qz,$$  \hspace{1cm} (1)

where the orientation of $k$ in the $x, y$ plane is not fixed, $2\pi/k$ is the period of the sinusoidal domain structure, $q = \pi/l$, and $l$ is the film thickness. To find the desired free energy, $F(a, p)$, one has to find the elastic strains and the non-ferroelectric polarization $P_\perp = (P_x, P_y)$ as functions of $a$ and $p$ to present the total free energy as a function of $a$ and $p$ only. The total free energy contains contributions of the ferroelectric film, of the substrate and of the electrode. In principle, it should also contain a contribution of the voltage source but we consider here a short-circuited system and are not concerned with the latter contribution.

When calculating elastic strains in the ferroelectric which accompany the inhomogeneous polarization forming the sinusoidal domain structure, we follow the same philosophy as in our previous work. In principle, when calculating these strains the inhomogeneous strains in the substrate should be taken into account. However, it is well known that they propagate into the substrate for about the same distances as the scale of inhomogeneity in the film $(x, y)$ plane. In our case, these inhomogeneities are due to the domain structure, i.e. this scale is the period of the domain structure. Then, it is convenient to consider relatively thick films since the period of the domain structure is relatively small, specifically, it is much less than the film thickness, Fig. 2. The contribution of the substrate is its elastic energy which, as we have mentioned above, is concentrated within a volume which is much smaller than the film volume, as defined by a small factor $q/k = \pi/kl \ll 1$. Another convenience of the thick film limit is that it is possible to disregard the boundary conditions for the inhomogeneous parts of elastic strains and stresses at the surfaces of the
FIG. 2: (color online) Schematic of the ferroelectric film on the misfit substrate at the onset of sinusoidal polarization wave. The elastic coupling to the substrate allows inhomogeneous deformations but prohibits homogeneous strains in plane of the film.

ferroelectric. Indeed, if we obtain a solution, which does not satisfy the boundary conditions, we can find corrections to such a solution in a way that is conventionally used in the elasticity theory. First, we apply the external forces to the surfaces, which are necessary to meet the boundary conditions with the strains corresponding to our solution making this solution correct. Second, we apply forces opposite to the previous ones and find the strains produced by the new forces. These strains provide the correction to the original solution we were looking for. Once more, it is sufficient to observe that in our case the external forces have the period of the domain structure to understand that the elastic energy associated with the corrections necessary to satisfy the boundary conditions can be neglected quite similarly to the elastic energy of the substrate. Another convenience of thick film approximation is given by the possibility to neglect those terms in the LGD free energy, which describe the electrostriction but contain components of non-ferroelectric polarization. According to Refs.6,8,

\[ \mathbf{P}_\perp(x, y, z) = (k/k)a_\perp \sin kr \sin qz, \] (2)

where \( a_\perp \approx aq/k, \frac{q}{k} = \pi/kl \ll 1 \). The electrostriction terms in the LGD free energy with non-ferroelectric components of polarization may contain the ferroelectric component, like \( P_x P_z u_{xz} \), or may not contain them, like in the term \( P_x P_y u_{xy} \). In both cases, they contribute to \( a^4 \) and \( p^2 a^2 \) terms in the free energy depending on \( a \) and \( p \). In the first case, this contribution is proportional to \( (q/k)^2 \) and in the second to \( (q/k)^4 \). Since there are also the terms \( a^4, p^2 a^2 \) that do not contain the small factor \( q/k \), the contribution of these terms can be neglected.

Taking this into account, we write down the LGD free energy in the form:

\[ F(P, u_{ik}) = F_1(P) + F_2(u_{ik}) + F_3(P, u_{ik}), \] (3)

where

\[ F_1(P) = \frac{A}{2} P_z^2 + \frac{B}{4} P_z^4 + \frac{1}{2} G (\nabla_\perp P_z)^2 + \frac{1}{2} \kappa P_{zz}^2 + \frac{A_\perp}{2} P_\perp^2, \] (4)

\[ F_2(u_{ik}) = \frac{1}{2} \lambda_1 \left( u_{xx}^2 + u_{yy}^2 + u_{zz}^2 \right) + \lambda_2 \left( u_{xx} u_{yy} + u_{xx} u_{zz} + u_{zz} u_{yy} \right) + 2 \mu (u_{zy}^2 + u_{zx}^2 + u_{xz}^2), \] (5)

\[ F_3(P, u_{ik}) = q_{11} u_{zz} P_z^2 + q_{12} (u_{xx} + u_{yy}) P_z^2. \] (6)

Here, \( q_{11(12)} \) are the standard piezo-electric coefficients that should not be confused with the parameter \( q \) defining the transversal profile of the polarization wave [1]. In Eq. [3], \( A = \gamma (T - T_c), B, G = \text{const}, \nabla_\perp (\partial_x, \partial_y) \) the gradient in the plane of the film, \( P_{zz} \) is the non-ferroelectric (‘base’) part of the polarization perpendicular to the electrodes[14], \( A_\perp > 0 \). Following Refs.6,8, we have neglected a term with the gradient in \( z \)-direction since it is much smaller than the one in plane of the film, \( \partial_z \ll \nabla_\perp \). It is worth mentioning that we have not included the energy of the electric
field into the LGD free energy. The reason is that we shall use it to write down the constituent equations only. We shall eliminate \( u_{ik}, \ P_\perp, \ \rho_k \) as well the electric field components from the system of constituent electrostatics equations to obtain two coupled equations of state for \( a \) and \( p \). We shall obtain \( F(p,a) \) from the resulting equations. This is possible because of the thick films approximation. The most straightforward method to obtain \( F(p,a) \) would be to substitute Eq.(1) into Eq.(3) supplemented by the electric field energy and to integrate over the film volume. In general, the result would not be the same as the one obtained from the constituent equations because of approximate character of Eq.(1). However, for \( q = \pi/l \) the two results coincide and that makes it possible to use a more convenient method of the constituent equations.

### III. CONSTITUENT EQUATIONS

For the polarization components one has:

\[
AP_z + BP_z^3 = G \nabla^2 P_z + 2q_{11} P_z u_{zz} + 2q_{12} P_z (u_{xx} + u_{yy}) = E_z,
\]

\[
P_{hz} = \kappa E_z,\]

\[
P_\perp = A_1 E_\perp.
\]

Before writing down the equations for the strain, we shall eliminate the electric field from the above three equations. Assuming Eq.(1) for \( P_z \), Eq.(2) for \( P_\perp \) and putting\(^a\):

\[
E_{0z} = E_0 + E_z^k \cos k \mathbf{r} \cos qz, \quad E_\perp = (k/k) E_\perp^k \sin k \mathbf{r} \sin qz
\]

we can replace Eqs.(7), (9) with

\[
A p + [BP_z^3 + 2q_{11} P_z u_{zz} + 2q_{12} P_z u_{\perp\perp}]_{\text{hom}} = E_0,
\]

\[
(A + Gk^2) a + [BP_z^3 + 2q_{11} P_z u_{zz} + 2q_{12} P_z u_{\perp\perp}]_{\text{cc}} = E_z^k,
\]

\[
A_1 a = E_z^k,
\]

where \( u_{\perp\perp} = u_{xx} + u_{yy} \), \([\ldots]\)_\text{hom} and \([\ldots]\)_\text{cc} denote the homogeneous part \((k = 0)\) and the part proportional to \( \cos k \mathbf{r} \cos qz \) of the expression in the brackets, correspondingly. Of course, as a result of this replacement, a part of the l.h.s. of Eq.(7) is lost but it corresponds to the higher harmonics of the sinusoidal distribution of the polarization and these harmonics can be neglected close to the transition\(^6,13\).

The homogeneous part of the electric field \( E_{z0} \) can be calculated as, e.g., in Ref.\(^\text{5}\) yielding for the short-circuited case

\[
E_{z0} = -\frac{4\pi d}{\varepsilon_0 d + \varepsilon_e l} p,
\]

where \( d \) is the thickness of the dead layer and \( \varepsilon_e \) its dielectric constant. Recall that real electrodes have finite albeit small (Thomas-Fermi) screening length \( \lambda \), which is completely analogous\(^5\) to a presence of the ‘dead’ non-ferroelectric layers at the interface with thickness \( d/2 = \lambda \). Using Eq. (13), the equation (11) gets the form:

\[
A_1 p + [BP_z^3 + 2q_{11} P_z u_{zz} + 2q_{12} P_z (u_{xx} + u_{yy})]_{\text{hom}} = 0.
\]

where

\[
A_1 = A + \frac{4\pi d}{\varepsilon_0 d + \varepsilon_e l} \approx A + \frac{4\pi d}{\varepsilon_e l},
\]

since usually the dead layer is very thin, \( \varepsilon_0 d \ll \varepsilon_e l \). To transform Eqs. (12) we use the electrostatics equation,

\[
\text{div} \mathbf{D} = 0,
\]

where \( \mathbf{D} \) is the dielectric displacement, firstly for the ferroelectric material, taking into account that \( \mathbf{D} = (\varepsilon_\perp E_\perp, \ \varepsilon_b E_z + 4\pi P_z) \), where \( \varepsilon_\perp = 1 + 4\pi/A_\perp \), and \( \varepsilon_b = 1 + 4\pi/\kappa \) is the ‘base’ non-critical dielectric constant\(^6,14\), and together with the equation \( \text{curl} \mathbf{E} = 0 \), we find that
$$E^k_z = -\frac{4\pi q^2}{\varepsilon_\perp k^2}a.$$  

Substituting (18) into the equation for the amplitude of the ‘polarization wave’ $a$, Eq. (12), we rewrite the latter as:

$$\left( A + Gk^2 + \frac{4\pi q^2}{\varepsilon_\perp k^2} \right) a + \left[ BP_z^3 + 2q_{11} P_z u_{zz} + 2q_{12} P_z u_{\perp\perp} \right]_{cc} = 0. \quad (19)$$

The nontrivial solution of the above equation first appears when the coefficient in the first term in round brackets before the amplitude $a$ in the above equation first crosses zero, i.e. when $A = \left[ -Gk^2 - 4\pi q^2 / (\varepsilon_\perp k^2) \right]_{\max}$. That takes place at some $A < 0$, so upon lowering temperature at constant thickness the transition into domain state occurs somewhat below the bulk critical temperature $T_c$, in other words. The transition for varying thickness of the film at some constant temperature $T < T_c$ takes place when the thickness exceeds some critical value. Therefore, the first nontrivial solution appears for the ‘polarization wave’ with the wave number $k$ that minimizes the sum $Gk^2 + 4\pi q^2 / (\varepsilon_\perp k^2)$, so that (recall that $q = \pi / l$)

$$\frac{4\pi q^2}{\varepsilon_\perp k^2} = Gk^2, \quad k = \left( \frac{4\pi q^2}{\varepsilon_\perp G} \right)^{1/4} = \left( \frac{4\pi^3}{\varepsilon_\perp G l^2} \right)^{1/4}. \quad (20)$$

We can now rewrite Eq. (12) as the homogeneous one:

$$A_2 a + \left[ BP_z^3 + 2q_{11} P_z u_{zz} + 2q_{12} P_z u_{\perp\perp} \right]_{cc} = 0, \quad (21)$$

where

$$A_2 = A + 2Gk^2. \quad (22)$$

It is seen from Eq. (6) that the only source of elastic stresses and strains is $P_z^2 (x, y, z)$ in our approximation. Since

$$P_z^2 = p^2 + 2pa \cos kr \cos qz + \frac{a^2}{4} \left( 1 + \cos 2qz + \cos 2kr + \cos 2qz \right), \quad (23)$$

we should expect that

$$u_{zz} = u_{zz}^{(0)} + u_{zz}^{(1)} \cos 2qz + u_{zz}^{(2)} \cos kr \cos qz + u_{zz}^{(3)} \cos 2kr + u_{zz}^{(4)} \cos 2qz, \quad (24)$$

while for $u_{xx}$, $u_{yy}$, and for $u_{\perp\perp}$ we shall have similar formulas with the homogeneous part (first term in the above expression) absent because of the substrate. The superscripts $(0) - (4)$ denote contributions with different types of the coordinate dependencies as defined by Eq. (24). Below, we use the same superscripts for both the coefficients and the functions.

Substituting Eqs. (23), (24) and analogous equations for $u_{xx}$ and $u_{yy}$ into Eqs. (15), (21), we find:

$$A_1 p + \left[ BP_z^3 \right]_{\text{hom}} + 2q_{11} \left( pu_{zz}^{(0)} + \frac{a u_{zz}^{(2)}}{4} \right) + q_{12} a \frac{u_{zz}^{(2)}}{2} = 0, \quad (25)$$

$$A_2 a + \left[ BP_z^3 \right]_{cc} + 2q_{11} \left[ pu_{zz}^{(2)} + a \left( u_{zz}^{(0)} + \frac{u_{zz}^{(1)} + u_{zz}^{(3)}}{2} + \frac{u_{zz}^{(4)}}{4} \right) \right] + 2q_{12} \left[ pu_{\perp\perp}^{(2)} + a \left( \frac{u_{\perp\perp}^{(1)} + u_{\perp\perp}^{(3)}}{2} + \frac{u_{\perp\perp}^{(4)}}{4} \right) \right] = 0. \quad (26)$$

We shall calculate the values $u_{ik}^{(j)}$ in the next Section by solving the elastic problem explicitly.

Importantly, the above equation of state (25) suggests that the film would tend to transform into a single domain (SD) state with $p \neq 0$ and $a = 0$ at temperature $T_c^{\text{SD}}$ such that $A_1 = 0$ or, in other words,

$$A(T_c^{\text{SD}}) = -4\pi d / (\varepsilon l) \quad (27)$$

The second equation of state (26) yields a transition into a domain state ($p = 0$ and $a \neq 0$) at the temperature $T_d$ such that $A_2 = 0$ or

$$A(T_d) = -2Gk^2 = -4 \left( \frac{\pi^3 G}{\varepsilon_\perp} \right)^{1/2} \frac{1}{l}. \quad (28)$$
Recall that in the present case, corresponding to \( \text{BaTiO}_3/\text{SrRuO}_3/\text{SrTiO}_3 \),
\[
\frac{4\pi d}{\epsilon_e} > 2Gk^2 \sim \frac{4\pi d_{at}}{\epsilon_{1/2}} \tag{29}
\]
where \( d_{at} = \sqrt{\pi G} \approx 1\,\text{Å} \) is the small ‘atomic’ length scale \( (G = 0.3\,\text{Å}^2 \text{ for } \text{BaTiO}_3) \). The above relation means that the paraphase gives way to the domain phase, with \( a \neq 0 \), thus preventing it from reaching the temperature \( T_d \) where it could have transformed into a single domain state. Obviously, same is true of the phase transformations in the film as a function of thickness at constant temperature. There, one can introduce the critical thickness for domains, \( l_d \), where \( A = -2Gk^2 = -4 \left( \frac{\pi^3 G}{\epsilon_{1/2}} \right) \left( \frac{1}{l_d} \right) \), \( \tag{30} \)
and the ‘critical thickness for the single domain state’ \( l_{SD}^c \), such that
\[
A = -4\pi d/ (\epsilon_{SD}^c) \tag{31}\]
These introduced critical thicknesses and temperatures are discussed in detail below in Sec. VI.

IV. ELASTIC PROBLEM

Using Eqs. (5),(6), we obtain for the diagonal components of the elastic stress tensor:
\[
\sigma_{xx} = \lambda_1 u_{xx} + \lambda_2 (u_{yy} + u_{zz}) + q_{12} P^2_z, \tag{32}
\]
\[
\sigma_{yy} = \lambda_1 u_{yy} + \lambda_2 (u_{xx} + u_{zz}) + q_{12} P^2_z, \tag{33}
\]
\[
\sigma_{zz} = \lambda_1 u_{zz} + \lambda_2 (u_{xx} + u_{yy}) + q_{11} P^2_z, \tag{34}
\]
and formulas of the type
\[
\sigma_{xy} = 2\mu u_{xy}, \tag{35}
\]
for the off-diagonal components.

We have already mentioned that the only \( u_{ik} \) component which has a non-zero homogeneous part is \( u_{zz} \). This part is easily found from the condition at the free surface: \( \sigma_{zz} = 0 \) at \( z = l/2 \). From Eq. (34), one finds:
\[
u_{zz}^{(0)} = \frac{q_{11}}{\lambda_1} \left[ P^2_z \right]_{\text{hom}} = -\frac{q_{11}}{\lambda_1} \left( p^2 + \frac{a^2}{4} \right). \tag{36} \]

For the parts depending on \( z \) only, the equations of elastic equilibrium take the form:
\[
\partial \sigma_{zz}^{(1)} / \partial z = 0, \tag{37}
\]
i.e. \( \sigma_{zz}^{(1)} = \text{const} = 0 \) since it should vanish at the free surface \( (z = l/2) \). Therefore, Eqs. (34), (23) yield
\[
u_{zz}^{(1)} = -q_{11} a^2 / (4\lambda_1), \tag{38} \]
and
\[
u_{xx}^{(1)} = \nu_{yy}^{(1)} = 0, \tag{39} \]
because of the Saint-Venant’s elastic compatibility conditions for \( z \)-only dependent strains.

When solving the rest of the elastic problem, we shall use the small parameter \( q/k \ll 1 \). This allows us to neglect the derivatives with respect to \( z \): formally, \( \partial / \partial z \ll \partial / \partial x, \partial / \partial y \). As a result, the equations of the elastic equilibrium acquire the form:
\[
\frac{\partial \sigma_{zz}^{(2-4)}}{\partial x} + \frac{\partial \sigma_{xy}^{(2-4)}}{\partial y} = \frac{\partial \sigma_{xz}^{(2-4)}}{\partial y} + \frac{\partial \sigma_{yz}^{(2-4)}}{\partial x} = \frac{\partial \sigma_{yy}^{(2-4)}}{\partial y} + \frac{\partial \sigma_{yx}^{(2-4)}}{\partial x} = 0, \tag{40}
\]
where the superscripts \((2-4)\) denote the part of stresses that is due to the three last terms in Eq. [23], which we denote as \(P_z^{2(2-4)}\). Explicitly,

\[
\lambda_1 \frac{\partial^2 u^2_x}{\partial x^2} + \lambda_2 \frac{\partial^2 u^2_y}{\partial y \partial x} + \mu \frac{\partial^2 u^2_x}{\partial y^2} + \mu \frac{\partial^2 u^2_y}{\partial y \partial x} + q_{12} \frac{\partial P_z^{2(2-4)}}{\partial x} = 0, \tag{41}
\]

\[
\mu \frac{\partial^2 u^2_z}{\partial y^2} + \mu \frac{\partial^2 u^2_x}{\partial x^2} = 0, \tag{42}
\]

\[
\lambda_1 \frac{\partial^2 u^2_y}{\partial y^2} + \lambda_2 \frac{\partial^2 u^2_y}{\partial y \partial x} + \mu \frac{\partial u^2_x}{\partial x} + \mu \frac{\partial^2 u^2_y}{\partial x^2} + q_{12} \frac{\partial P_z^{2(2-4)}}{\partial y} = 0. \tag{43}
\]

Analogously to the isotropic case\(^{13}\), we shall put the conditions \(u^{2(4)} = 0\) that satisfy Eq. [42] but not, of course, the boundary conditions. The latter is not important in our approximation, as we argued above. Therefore, we conclude that

\[
u^{2}_zz = u^{3}_zz = u^{4}_zz = 0, \tag{44}
\]

and we are left with only two equations to solve. It is convenient to solve them separately for \((2)\) and \((3,4)\) parts, since they correspond to different spatial harmonics.

Simplifying the remaining equations \([41,43]\), we obtain:

\[
\lambda_1 \frac{\partial^2 u^{2(4)}_x}{\partial x^2} + (\lambda_2 + \mu) \frac{\partial^2 u^{2(4)}_y}{\partial y \partial x} + \mu \frac{\partial^2 u^{2(4)}_x}{\partial y^2} + q_{12} \frac{\partial P_z^{2(2-4)}}{\partial x} = 0, \tag{45}
\]

\[
\lambda_1 \frac{\partial^2 u^{2(4)}_y}{\partial y^2} + (\lambda_2 + \mu) \frac{\partial^2 u^{2(4)}_x}{\partial y \partial x} + \mu \frac{\partial^2 u^{2(4)}_y}{\partial x^2} + q_{12} \frac{\partial P_z^{2(2-4)}}{\partial y} = 0. \tag{46}
\]

For the terms \(u^{2}\), we have \(P_z^{2(2)} \propto \cos kr, \quad \partial_x(y)P_z^{2} \propto -k_x(y) \sin kr\), meaning that \(u_{x(y)} \propto \sin kr, \quad \partial^2 u_i / \partial x^2 = -k_x^2 u_i\), etc. Then,

\[
\lambda_1 k_x^2 u^{2}_x + (\lambda_2 + \mu) k_x k_y u^{2}_y + \mu k_x^2 u^{2}_x + q_{12} k_x 2pa = 0, \tag{47}
\]

\[
\lambda_1 k_y^2 u^{2}_y + (\lambda_2 + \mu) k_x k_y u^{2}_x + \mu k_y^2 u^{2}_y + q_{12} k_y 2pa = 0. \tag{48}
\]

The terms \(u^{3,4}\) correspond to higher spatial harmonics in Eq. [23], but they should be taken into account since they contribute to the terms with the main harmonic in the constituent equation [20]. Since for this part \(P_z^{2} \propto \cos 2kr\), we obtain a slightly different set of equations:

\[
\lambda_1 k_x^2 u^{3,4}_x + (\lambda_2 + \mu) k_x k_y u^{3,4}_y + \mu k_x^2 u^{3,4}_x + q_{12} k_x a^2 / 8 = 0, \tag{49}
\]

\[
\lambda_1 k_y^2 u^{3,4}_y + (\lambda_2 + \mu) k_x k_y u^{3,4}_x + \mu k_y^2 u^{3,4}_y + q_{12} k_y a^2 / 8 = 0. \tag{50}
\]

Note that for the constituent equations we need the combinations:

\[
u^{2}_{2} = u^{(2)}_{xx} + u^{(2)}_{yy} = k_x u^{(2)}_x + k_y u^{(2)}_y, \]

\[
u^{3,4}_{1-1} = u^{(3,4)}_{xx} + u^{(3,4)}_{yy} = 2k_x u^{(3,4)}_x + 2k_y u^{(3,4)}_y.
\]

We find:

\[
u^{2}_{1-1} = -2q_{12} ap f (\theta), \tag{51}
\]

where \(\theta\) is defined by \(k_x = k \cos \theta, \quad k_y = k \sin \theta\) and the function \(f (\theta)\) is
\[ f(\theta) = \frac{(\lambda_1 - \lambda_2) \sin^2 2\theta + 2\mu \cos^2 2\theta}{(\lambda_1 + \lambda_2 + 2\mu)(\lambda_1 - \lambda_2) \sin^2 2\theta + 4\lambda_1 \mu \cos^2 2\theta}. \]  

(52)

For terms corresponding to \( \cos 2k r \) and \( \cos 2q z \), we obtain:

\[ u_{11}^{(3)} = u_{11}^{(4)} = -a^2 \frac{q^2}{3} f(\theta). \]

(53)

Using the results of solution of the elastic problem, Eqs. (36), (44), (51), (53), we can write Eqs. (25), (26) as

\[ A_1 p + \left( B - \frac{2q^2}{\lambda_1} \right) p^3 + pa^2 \frac{3}{4} \left[ B - \frac{4}{3} \left( \frac{q^2}{2\lambda_1} + q_{12}^2 f(\theta) \right) \right] = 0 \]

(54)

\[ A_2 a + a^3 \frac{9}{16} \left( B - \frac{4}{3} \left[ \frac{q_{11}^2}{\lambda_1} + \frac{q_{12}^2}{2} f(\theta) \right] \right) + ap^2 \frac{3}{8} \left( B - \frac{4}{3} \left[ \frac{q_{11}^2}{2\lambda_1} + q_{12}^2 f(\theta) \right] \right) = 0. \]

(55)

From these two constituent equations corresponding to an extremum of the free energy, one can easily reconstruct the free energy:

\[ V^{-1} \tilde{F}(p, a) = \frac{A_1}{2} p^2 + \frac{A_2}{8} a^2 + \frac{\bar{B}}{4} p^4 + \frac{3B_1(\theta)}{8} a^2 p^2 + \frac{9B_2(\theta)}{256} a^4, \]

(56)

where

\[ \bar{B} = B - \frac{2q_{11}^2}{\lambda_1}, \quad B_1(\theta) = \bar{B} + \frac{4}{3} \left[ \frac{q_{11}^2}{\lambda_1} - q_{12}^2 f(\theta) \right], \quad B_2(\theta) = \bar{B} + \frac{2}{3} \left[ \frac{q_{11}^2}{\lambda_1} - q_{12}^2 f(\theta) \right]. \]

(57)

are the Landau coefficients before the quartic terms renormalized by the strain. The form of the free energy is the same as in the isotropic case\(^6,8,13\), but, importantly, the coefficients \( B_1 \) and \( B_2 \) depend on the orientation of the 'polarization wave' given by the angle \( \theta \).

V. ORIENTATION OF THE DOMAIN STRUCTURE

Consider the domain structure formed close to the paraelectric-ferroelectric transition. Although stability of the paraelectric phase is lost with respect to the polarization waves with the value of the \( k \) vector given by Eq. (20) and arbitrary orientation in the \( x - y \) plane, i.e. for any \( \theta \), the energy of sinusoidal domain structure depends on \( \theta \) and one has to find the ones corresponding to the equilibrium domain structure(s). Since we consider here one ‘polarization wave’ only, we study the competition between the stripe-type structures. In Sec. VII we shall show that the square (checkerboard) domain structure is unstable in perovskite crystals that we study here. The checkerboard structure could, in principle, be stable or metastable under some conditions on the material constants, but we are not aware of any experimental example of this type, so it will be premature to study such a hypothetical case.

Recall that we discuss the ferroelectric phase transition in a sample with short-circuited electrodes. Then, \( p = 0 \) in the ferroelectric phase at least not far from the phase transition, and the phase transition into the inhomogeneous domain phase occurs at \( A_2 = 0 \). The free energy is:

\[ V^{-1} \tilde{F}(p, a) = \frac{A_2}{8} a^2 + \frac{9B_2(\theta)}{256} a^4. \]

(58)

At a fixed \( \theta \), the minimum of this free energy is realized for

\[ a^2 = -\frac{16A_2}{9B_2(\theta)}, \]

(59)

with the corresponding free energy

\[ V^{-1} \tilde{F}_{\text{min}}(p, a) = -\frac{A_2^2}{9B_2(\theta)}. \]

(60)
We see that the equilibrium domain structure is realized for the angles \( \theta \) which minimize the function \( B_2(\theta) \) or, according to Eq. (57), maximize the function \( f(\theta) \). Let us find maxima of this function. It can be written in the form:

\[
 f(\theta) = \frac{2}{\lambda_1 + \lambda_2 + 2\mu} \left( 1 + \frac{r - c}{\tan^2 2\theta + c} \right),
\]

where

\[
 r = \frac{2\mu}{\lambda_1 - \lambda_2}, \quad c = \frac{4\lambda_1\mu}{(\lambda_1 + \lambda_2 + 2\mu)(\lambda_1 - \lambda_2)}.
\]

One sees that for \( r > c \) or

\[
 \lambda_2 + 2\mu > \lambda_1,
\]

the equilibrium domain structure corresponds to \( \theta_{eq} = 0, \pi/2 \) and

\[
 f(\theta)_{\text{max}} = f(0) = 1/\lambda_1,
\]

while in the opposite case \( \theta_{eq} = \pi/4, 3\pi/4 \) and

\[
 f(\theta)_{\text{max}} = f(\pi/4) = \frac{2}{\lambda_1 + \lambda_2 + 2\mu}. \tag{64}
\]

Throughout the present paper, we use the data for the material constants of BaTiO\(_3\) and PbTiO\(_3\) from Refs. 9,15–18 and of Pb(Zr\(_{0.5}\)Ti\(_{0.5}\))O\(_3\) from Ref. 22. We see that for BTO and PTO the condition of Eq. (63) is met and, therefore, the equilibrium 180° domain structure consists of stripes parallel (perpendicular) to the cubic axes, while the opposite inequality applies to PZT and the stripes make 45° with the cubic axes there. For BTO, our conclusion coincides with that of Dvorak and Janovec 19 who defined the equilibrium orientation of the 180° domain walls in BTO far from the phase transition. These authors were surprised by their conclusion about a very weak orientational dependence of the domain wall energy given that the experimental observations 20 showed a clearly preferable orientation, the same as suggested by the theory.

It follows from our results that the weak orientational dependence of the domain structure energy takes place in the sinusoidal regime too, and not only for BTO, but for all three perovskites we have made the numerical estimates for. Indeed, from Eq. (60) one sees that the orientational dependence of the domain structure energy comes from the function \( B_2(\theta)\). The maximum difference of values Eq. (57) for \( B_2(\theta) \) can be used to characterize the anizotropy of the domain wall energy:

\[
 \Delta B_2 = B_{2\text{max}} - B_{2\text{min}} = \frac{2}{3} q^2 \left| f(0) - f(\pi/4) \right| = \frac{4}{3} q^2 |\lambda_1 - \lambda_2 - 2\mu| \frac{|\lambda_1 - \lambda_2 + 2\mu|}{\lambda_1 + \lambda_2 + 2\mu}. \tag{65}
\]

We found \( \Delta B_2/B_2 \sim 4 \times 10^{-3} \) for BTO, much smaller anizotropy \( \sim 3 \times 10^{-4} \) for PTO, and even smaller one for PZT where \( \Delta B_2/B_2 \sim 4 \times 10^{-5} \) (we have used the parameters listed in the footnote 22). Such a weak angular dependence of the domain structure energy is in accordance with phase field results of Ref. 16 that shows domain walls mainly with thermodynamically favorable orientations but also with strong deviations from them.

VI. LOSS OF STABILITY OF A SINGLE DOMAIN STATE

It is convenient to study the loss of stability of the single domain state with respect to formation of a domain structure using Eq. (50). With this, we mean the loss of stability with respect to an arbitrarily small ‘polarization waves’ so that the original single domain state may be, in principle, either stable or metastable. Specifically, in our case, when Eq. (29) is valid, this state is metastable 16.

A solution of the equations,

\[
 \partial \tilde{F}/\partial p = 0, \quad \partial \tilde{F}/\partial a = 0, \tag{66}
\]

corresponding to a single domain state \( (p \neq 0, a = 0) \) is possible only if \( a_1 < 0 \) with \( p_{extr}^2 = -a_1/\tilde{B} \), where the subscript stands for the ‘extremum’. This extremum is a minimum (which is relative in our case) if

\[
 \partial^2 \tilde{F}/\partial p^2 > 0, \quad \partial^2 \tilde{F}/\partial a^2 > 0, \tag{67}
\]
FIG. 3: Schematic of the sinusoidal domain structure in (a) BTO and PTO, and (b) PZT. While the stripes are oriented along the crystal axes in case (a), in case (b) the stripes are at 45 degrees with respect to cubic axes, the difference being due to an opposite sign of elastic anisotropy in those two cases.
at the point $p = p_{\text{extr}}$, $a = 0$ given that $\partial^2 \tilde{F} / \partial p \partial a$ is evidently zero at this point. The first inequality in Eq. (67) is obviously valid for $A_1 < 0$, while the validity of the second is not immediately evident.

We find from (56):

$$4 \left( \frac{\partial^2 \tilde{F}}{\partial a^2} \right)_{a=0, p=p_{\text{extr}}} = A_2 + 3B_1 (\theta) p_{\text{extr}}^2 = A_2 - 3A_1 - 3A_1 \left( B_1 (\theta) - \tilde{B} \right) / \tilde{B},$$

(68)

From the condition $\left( \frac{\partial^2 \tilde{F}}{\partial a^2} \right)_{a=0, p=p_{\text{extr}}} = 0$, we obtain the value of $A$ corresponding to a loss of stability of the single domain state with respect to appearance of a polarization wave with a given orientation, $A_{pw} (\theta)$. It is convenient to present it in the form:

$$A_{pw} (\theta) = -\frac{6\pi d}{\varepsilon_b d + \varepsilon_e l} + Gk^2 + \left( \frac{4\pi d}{\varepsilon_b d + \varepsilon_e l} - 2Gk^2 \right) \beta (\theta),$$

(69)

where

$$\beta (\theta) = \frac{q_{11}^2 - q_{12}^2 f (\theta) \lambda_1}{B \lambda_1 + 2 (q_{11}^2 - q_{12}^2 f (\theta) \lambda_1)}.$$

(70)

The last term in Eq. (69) is the result of the polarization-strain coupling while the first two present the prior case without this coupling. According to Eq. (67), the corresponding single domain state will be (meta)stable at low temperatures such that $A < \min A_{pw} (\theta)$.

The actual loss of stability of the single domain state corresponds to the minimum of $A_{pw} (\theta)$. We have seen in Sec. [X] that for perovskites the angular dependencies are very weak and we can neglect it putting $f \lambda_1 = 1$. Then

$$\beta = \frac{q_{11}^2 - q_{12}^2}{B \lambda_1 + 2 (q_{11}^2 - q_{12}^2)}$$

(71)

Since in BTO, PTO, and PZT $q_{11}^2 > q_{12}^2$, the last term in Eq. (69) is positive and, therefore, the region of metastability of the single domain state in these systems is broader than according to and, in apparent accordance with. However, there are serious reservations. First of all, the effect is not very spectacular. Indeed, the factor $\beta$ in the last term of Eq. (69) is always less than one half, $\beta < 1/2$, approaching that value when $q_{11}^2 - q_{12}^2 \to \infty$. Therefore,

$$A_{pw} < -\frac{4\pi d}{\varepsilon_b d + \varepsilon_e l},$$

(72)

where the r.h.s. corresponds to a very strong strain coupling. Recall that $A = -4\pi d / (\varepsilon_b d + \varepsilon_e l)$, or $A_1 = 0$, corresponds to what was calculated in several papers as a `critical thickness of single-domain ferroelectricity' $l_{\text{SD}}^{\text{cr}}$, Eq. (61).

To get the opposite limit of a weak strain coupling for $A_{pw} (0)$, we put $q_{11}^2 - q_{12}^2 = 0$ and neglect $Gk^2$, as Pertsev and Kohlstedt did. We see that

$$-\frac{6\pi d}{\varepsilon_b d + \varepsilon_e l} < A_{pw} (0) < -\frac{4\pi d}{\varepsilon_b d + \varepsilon_e l},$$

(73)

i.e. because of account for the polarization-strain coupling the value of $A_{pw} (0)$ changes always by less than 1.5 times. In usual situation when $\varepsilon_b d < \varepsilon_e l$ this is also the interval of change of the thickness corresponding to absolute loss of stability of the single domain state at a fixed temperature.

The above moderate, less than 50%, range of change is in striking disagreement with a statement by Pertsev and Kohlstedt who claimed a more than an order of magnitude change by nullifying the electrostrictive constants. They do not report the details of their procedure, but it is clear from the rest of the paper that their suggestion of putting the electrostrictive constants to zero implied changes in the coefficients of the LGD free energy that should have been renormalized by the misfit strains. Evidently, it has nothing to do with the effects of the polarization-strain coupling omitted in Refs., since this renormalization is automatically taken into account there, while the effect of misfit strain on LGD coefficients was apparently neglected in a gedanken exercise performed in Ref.

Specifically, we find that for the perovskites BTO and PTO $\beta = 0.4$, while in PZT this parameter is 0.1, i.e. four times smaller. We see that BTO and PTO are similar and closer to the limit $q_{11}^2 - q_{12}^2 \to \infty$, $\beta = 0.5$, i.e. the point of loss of stability of single domain state is quite close in these materials to the `critical thickness of single-domain ferroelectricity' studied in Ref. and elsewhere. However, the latter does not have any practical importance because
FIG. 4: (color online) Regions of (meta)stability of the single domain and polydomain states in the ferroelectric film as a function of temperature $T$ at fixed thickness $l$ (top) and as a function of the film thickness $l$ at fixed $T$ (bottom). Upon lowering the temperature at a fixed thickness $l$ (top panel), the paraphase gives way to domains that are stable at all temperatures $T < T_d$, where $T_d$ is below the critical temperature of the bulk ferroelectric transition $T_c$. The single domain (SD) state is metastable at low temperatures $T < T_{SDms}$, when a strain coupling is neglected. The strain coupling shifts the boundary of metastability towards so-called critical temperature for a single domain state, $T_{SDc}$, as marked by the vertical arrows for the perovskites in question. The phase behavior of the films as a function of their thickness $l$ at fixed temperature (bottom) is qualitatively similar. Very thin films are in a paraelectric phase that is replaced by domains at larger thicknesses $l > l_d$. The single domain state is metastable at thicknesses $l > l_{SDms}$ and becomes suitable for memory applications at even larger (yet to be determined) thicknesses when the life time of the metastable state becomes sufficiently large. Strain coupling may extend the boundary of the metastability down to the so-called ‘critical thickness for SD ferroelectric state’ $l_{SDc}$. The films with FE memory would be somewhere at larger thicknesses in the metastable region, as marked on the diagrams.

if one fixes the temperature and reduces the film thickness starting with a monodomain ferroelectric state at low temperatures or large thicknesses, that state will give way to domains before the thickness determined by the limit of the single domain state stability is reached. The fact of the matter is that the single domain state is metastable, it may have a large lifetime at low temperatures and large film thicknesses but this lifetime goes essentially to zero (to ‘atomically’ short times) when the above mentioned temperature or thickness are reached.

Importantly, it follows from Eq. (69) that if $q_{11}^2 < q_{12}^2$, the account for the inhomogeneous strains shrinks the region of metastability of the single domain state. This shows that contrary to the claim by Pertsev and Kohlstedt there is no general physical phenomenon such as stabilization of a single domain state because of inhomogeneous strains accompanying formation of domains. This may seem surprising, because solids are known to ‘dislike’ the inhomogeneous strains (free energy usually goes up). Moreover, the expectation of Pertsev and Kohlstedt is justified for a free-standing film, at least for elastically isotropic solids. But it is not certain for a film on substrate considered both by them and in the present work. To explain the physical reason, we recall that the coupling with strain renormalizes the coefficients before fourth-order terms in the LGD free energy, in our case we mean the coefficients before $p^4$ and $p^2a^2$ terms. Then, one has to take into account that the homogeneous strains in the plane of the substrate are not possible while inhomogeneous ones are. Both homogeneous and inhomogeneous polarization create homogeneous strain but to a different extent see Eqs. (56) and (58).
FIG. 5: (color online) The phase diagram for BaTiO$_3$/SrRuO$_3$/SrTiO$_3$ films in the plane temperature-thickness. The line marked $l_d$ delineates the para- and domain phases, while the one marked $l_{ms}$ marks the boundary of the metastability regions of the single domain state calculated for BTO accounting for strain coupling, Eq. (69). The broken line at larger thicknesses from $l_{ms}$ marks the metastable region calculated without accounting for the strain coupling, Eq. (76). The films with FE memory would be somewhere at larger thicknesses in the metastable region.

inhomogeneous polarization only. The out-of-plane and in-plane strains couple with the ferroelectric polarization $P_z$ by electrostriction terms with different coefficients and the final result of renormalization of the coefficient of $p^2 a^2$ term is due to several contributions and it is not clear upfront. It should be obtained by a consistent analysis, as it has been done above. No reason is seen to discard the possibility that the inequality $q_{12}^2 < q_{11}^2$ can be realized in some systems, and one cannot exclude, at least for the moment, the possibility of favoring the multidomain state by the polarization-strain coupling. Interestingly enough, this favoring may be very strong: according to Eq. (69), the increase of the region of absolute instability of single domain state becomes infinite when $q_{12}^2 - q_{11}^2$ tends to $B \lambda_1^2/2$ from below.

The ‘phase diagrams’ for the epitaxial FE films on a misfit substrate are plotted in Figs. 4, 5. The boundaries of paraelectric phase, domains, and metastable single domain region for BaTiO$_3$/SrRuO$_3$/SrTiO$_3$ system are shown in the temperature-film thickness ($T - l$) plane in Fig 5. They are found from the conditions that we discussed above and write down here for a reference:

$$A = -2Gk^2 = -4 \left( \frac{\pi^3 G}{\epsilon_{\perp}} \right)^{1/2} \frac{1}{l_d},$$

$$A = -\frac{4\pi d}{\epsilon_c l_{SD}^2},$$

$$A = -\frac{6\pi d}{\epsilon_c l_{SD}^{ms}},$$

where the Landau coefficient $A$ is evaluated for a given temperature $T$ of interest, $d/2 = \lambda = 0.8\AA$, $\epsilon_c = 8.45$ for
SrRuO$_4$ electrode, $G = 0.3\text{Å}^2$, and $\varepsilon_{\perp}$ the dielectric constant [see its definition below Eq.(17)] in the plane of the FE film has been found from the Landau coefficients. The last condition corresponds to $l = l_{ms}^{SD}$ found without accounting for the strain coupling. The critical line $T - l_{ms}^{SD}$ in the phase diagram, Fig.6, for BTO with an account for strain coupling has been found from Eq.(69). The arrows on Fig.6 show the evolution of the state at either $T = \text{const}$ or $l = \text{const}$.

One should understand that in both illustrations it is implied that the corresponding critical points have physical values as solutions to the conditions (??), or, equivalently, Eqs.(25),(27),(30),(31). Consider first the lowering of the temperature at a fixed thickness $l$ (Fig.4, top panel), where the paraphase transforms into domain state below the temperature $T_d$ that is smaller than the critical temperature of the bulk ferroelectric transition $T_c$. We see that the single domain (SD) state would be metastable at low temperatures $T < T_{ms}^{SD}$ in the region overlapping with the domain state. Note that the $T_{ms}$ plotted in Fig.6 is found without accounting for the strain coupling. The strain coupling then shifts the boundary of metastability towards the so-called critical temperature for a single domain state $T_c^{SD}$ thus broadening the range of metastability of the SD state, as shown in Fig.4. The phase behavior of the films as a function of their thickness $l$ at fixed temperature (Fig.4, bottom panel) is qualitatively similar. Very thin films remain in a paraelectric phase that is replaced by the domains at large $l$ thicknesses (right to left on the phase diagram, Fig.5), but such transitions are preempted by the domain instability that sets in first. The single domain state is metastable at thicknesses $l > l_{ms}^{SD}$, and becomes suitable for memory applications at even larger (yet to be determined) thicknesses where its life time becomes sufficiently long.

VII. INSTABILITY OF THE CHECKERBOARD DOMAIN STRUCTURE

In the previous Sections, we have assumed that the domain structure is stripe-like by taking into account only one ‘polarization wave’. This and other possibilities have been studied by Chensky and Tarasenko who considered the uniaxial ferroelectric isotropic in the $x-y$ plane. Along with the stripe-like structure they discussed also the checkerboard and the hexagonal domain structures. The latter can be realized in the presence of an external field only which is not discussed in this paper. However, a checkerboard structure should be analyzed as an alternative to the stripe structure. In Ref.8, the authors stated that the checkerboard structure never realizes, although, surprisingly, there is no proof of this statement. In this Section, we shall show that this structure is indeed unstable for the isotropic case treated in Ref.8 and then show that this conclusion holds also when one explicitly takes into account the polarization-strain interaction, apart from mere renormalization of the LGD coefficients by the misfit strains.

Once again, we consider a short-circuited sample, i.e. the ferroelectric polarization is described by:

$$P_z = a_1 \cos k_1 r \cos q z + a_2 \cos k_2 r \cos q z,$$

where $k_1$ and $k_2$ are two noncollinear vectors whose modulus is given by Eq.(20) and whose (mutually orthogonal) directions remain unspecified for a moment, Fig.6.

A. Checkerboard domains without elastic coupling ($q_{11} = q_{22} = 0$)

In this case the solution for the fields is [cf. Eq.(11)]

$$E_z = E_z^{k_1} \cos k_1 r \cos q z + E_z^{k_2} \cos k_2 r \cos q z,$$

$$E_{x,y} = E_{x,y}^{k_1} \sin k_1 r \sin q z + E_{x,y}^{k_2} \sin k_2 r \sin q z,$$

with Eq.(18) still applicable to spatial harmonics (as follows from linearity of Maxwell equations) and the equation of state for the fundamental harmonics is the same as Eq.(12):

$$A_{2a_1} + [BP_{z,cc}^3] = E_z^{k_1},$$

where one retains the terms $[BP_{z,cc}^3] \propto \cos k_1 r \cos q z$ (symmetry dictates the analogous expressions for $a_2$). In the above equation,

$$P_z = (a_1 \cos k_1 r + a_2 \cos k_2 r)^3 \cos^3 q z = \frac{9}{16} (a_1^3 + 2a_1 a_2^2) \cos k_1 r \cos q z + \frac{9}{16} (a_2^3 + 2a_2 a_1^2) \cos k_2 r \cos q z + \ldots,$$

so that we obtain for the fundamental harmonic the following equations of state:

$$A_{2a_1} + \frac{9B}{16} (a_1^3 + 2a_1 a_2^2) = 0$$

(82)
FIG. 6: Schematic of the checkerboard domain structure. It is absolutely unstable in case of BaTiO$_3$, PbTiO$_3$, and Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$ typical perovskite ferroelectrics.

and the analogous equation for $a_2$. Since these equations are obtained from extremum of the free energy, $\partial \tilde{F}/\partial a_{1(2)} = 0$, we again restore the full free energy, accounting for the symmetric contribution by the $a_2$ harmonic:

$$V^{-1}\tilde{F} = \frac{A_2}{8} (a_1^2 + a_2^2) + \frac{9}{256} B (a_1^4 + a_2^4) + \frac{9}{64} Ba_1^2a_2^2.$$  (83)

The equations of state have the checkerboard solution:

$$a_1^2 = a_2^2 = -16A_2/27B.$$  (84)

Checking what type of extremum for the free energy is this solution,

$$\frac{\partial^2 F}{\partial a_1^2} \times \frac{\partial^2 F}{\partial a_2^2} - \left( \frac{\partial^2 F}{\partial a_1 \partial a_2} \right)^2 = -\frac{1}{12} A_2^2 < 0,$$

we see that the checkerboard solution is the maximum of the free energy for some directions in the $a_1, a_2$ plane and is absolutely unstable.

**B. Checkerboard domains with elastic coupling**

We have seen above that the elastic coupling renormalizes the fourth order coefficients in formulas like Eq. (83), reducing them by some amounts which are different for different coefficients. Thus, instead of Eq. (83), we will have:

$$V^{-1}\tilde{F} = \frac{A_2}{8} (a_1^2 + a_2^2) + \frac{9}{256} (B_{21}a_1^4 + B_{22}a_2^4) + \frac{9}{64} B_3a_1^2a_2^2.$$  (85)

where $B_{21}$ and $B_{22}$ are given by Eq. (57) for the corresponding angles and $B_3$ is a new coefficient which depends on the both angles and which can be, in principle, both positive and negative. Both from Eq. (57) and the cubic symmetry
one realizes that $B_{21} = B_{22} = B_2$. For what follows, it is important to mention that when $B_3$ is negative it cannot be of large absolute value, otherwise there will be directions in the $(a_1, a_2)$ plane along which the free energy diminishes without limits at large values of $a_1, a_2$ what means a global instability of the system. By putting $a_1 = a_2$, one sees from Eq. (80) that to avoid this instability the condition

$$B_2 + 2B_3 > 0$$

should be fulfilled. Another evident condition of the global stability is $B_2 > 0$.

The checkerboard solution is:

$$a_1^2 = a_2^2 = \frac{-16A_2}{9(B_2 + 2B_3)}.$$  

(87)

To analyze stability of this solution, we calculate the second derivatives

$$\frac{\partial^2 F}{\partial a_1^2} = \frac{\partial^2 F}{\partial a_2^2} = \frac{A_2}{4} + \frac{27}{64}B_2a_1^2(2) + \frac{9}{32}B_3a_2^2(1) = -\frac{1}{2}A\frac{B_2}{B_2 + 2B_3},$$

$$\frac{\partial^2 F}{\partial a_1 \partial a_2} = \frac{9}{16}B_3a_1a_2 = \pm \frac{A_2B_3}{B_2 + 2B_3}$$

we then find the discriminant

$$Z = \left(\frac{\partial^2 F}{\partial a_1^2}\right)\left(\frac{\partial^2 F}{\partial a_2^2}\right) - \left(\frac{\partial^2 F}{\partial a_1 \partial a_2}\right)^2 = \frac{1}{4}A^2\frac{B_2 - 2B_3}{B_2 + 2B_3}.$$  

(88)

Our further study is aimed at finding out if and when the condition of positiveness of $Z$, i.e. $B_2 > 2B_3$, is compatible with the two conditions of the global stability mentioned above. Thus we need formulas for the coefficients $B_2$ and $B_3$.

Turning to taking into account explicitly the polarization-strain coupling, we recall that in our approximation of sufficiently thick film the only source of the elastic strains is $P^2$. This function contains now a cross term stemming from

$$P^2 = (a_1 \cos k_1 r \cos qz + a_2 \cos k_2 r \cos qz)^2$$

$$= [a_1^2 + a_2^2 + 2a_1a_2 (\cos p^+ r + \cos p^- r) + a_1^2 \cos 2k_1 r + a_2^2 \cos 2k_2 r] \frac{1 + \cos 2qz}{4},$$  

(90)

where $p^\pm = k_1 \pm k_2$. Naturally, the components of the strain tensor will have terms depending on $\cos p^+ r$, $\cos p^- r$.

We will have for $u_{xx}$

$$u_{xx} = u^{(0)}_{xx} + u^{(1)}_{xx} \cos 2qz + u^p_{xx} \cos p^+ r + u^p_{xx} \cos p^- r + u^q_{xx} \cos p^+ r \cos 2qz$$

$$+ u^q_{xx} \cos p^- r \cos 2qz + u^{(3)}_{1,xx} \cos 2k_1 r + u^{(3)}_{2,xx} \cos 2k_2 r + \left(u^{(4)}_{1,xx} \cos 2k_1 r + u^{(4)}_{2,xx} \cos 2k_2 r\right) \cos 2qz,$$

and the analogous equations for $u_{yy}$ and $u_{zz}$.

From our previous experience, it becomes immediately clear that $u^p_{xx(yy)} = u^q_{xx(yy)}$, since the equations for those components do not depend on $z$ and we can now drop the indices $p$ and $q$ from the corresponding terms, leaving only $u^{(1)}_{xx(yy)}$. Also, due to the same reason as above, $u^{(1)}_{xx(yy)} = u^{(1)}_{xx(yy)} = 0$. Then, one can write:

$$u_{xx} = \left(u^+_{xx} \cos p^+ r + u^-_{xx} \cos p^- r\right) (1 + \cos 2qz)$$

$$+ u^{(3)}_{1,xx} \cos 2k_1 r + u^{(3)}_{2,xx} \cos 2k_2 r + \left(u^{(4)}_{1,xx} \cos 2k_1 r + u^{(4)}_{2,xx} \cos 2k_2 r\right) \cos 2qz.$$  

and a similar equation for $u_{yy}$. The ‘diagonal’ terms for the first (second) $k_{1(2)}$ harmonic are

$$u^{(3)}_{1(2),xx} + u^{(3)}_{1(2),yy} = u^{(4)}_{1(2),xx} + u^{(4)}_{1(2),yy} = -q_{12} \frac{a_{1(2)}^2 f(\theta_{1(2)})}{4}.$$
All the cross terms satisfy

\[
\lambda_1 \frac{\partial^2 u_x^\pm}{\partial x^2} + (\lambda_2 + \mu) \frac{\partial^2 u_y^\pm}{\partial y \partial x} + \mu \frac{\partial^2 u_y^\pm}{\partial y^2} + q_{12} \frac{\partial P_z^\pm}{\partial x} = 0, \tag{91}
\]

\[
\lambda_1 \frac{\partial^2 u_y^\pm}{\partial y^2} + (\lambda_2 + \mu) \frac{\partial^2 u_x^\pm}{\partial y \partial x} + \mu \frac{\partial^2 u_x^\pm}{\partial x^2} + q_{12} \frac{\partial P_z^\pm}{\partial y} = 0, \tag{92}
\]

where \( P_z^\pm = \frac{1}{2} a_1 a_2 \cos p^\pm \mathbf{r} \), in components \( u_x^\pm \propto \sin p^\pm \mathbf{r} \), \( \partial^2 u_i^\pm / \partial x^2 = - (p_x^\pm)^2 u_i^\pm \) etc.

\[
\lambda_1 p_x^\pm u_x^\pm + (\lambda_2 + \mu) p_y^\pm u_y^\pm + \mu p_x^\pm u_y^\pm + q_{12} p_x^\pm \frac{1}{2} a_1 a_2 = 0, \tag{93}
\]

\[
\lambda_1 p_y^\pm u_y^\pm + (\lambda_2 + \mu) p_y^\pm u_x^\pm + \mu p_y^\pm u_y^\pm + q_{12} p_y^\pm \frac{1}{2} a_1 a_2 = 0. \tag{94}
\]

Then,

\[
u_{xx}^\pm + u_{yy}^\pm = p_x^\pm u_x^\pm + p_y^\pm u_y^\pm = -q_{12} \frac{a_1 a_2}{2} f(\theta^\pm). \tag{95}\]

For \( u_{zz} \) we conclude, as above, that only \( u_{zz}^{(0)} \) and \( u_{zz}^{(1)} \) are non-zero and following the same reasoning as for the stripe phase, we obtain

\[
u_{zz}^{(0)} = u_{zz}^{(1)} = -\frac{q_{11}}{4 \lambda_1} (a_1^2 + a_2^2).\]

Having solved the elastic problem, we are now in a position to write down the constituent equations containing \( a_1 \) and \( a_2 \) only. To this end, we write two equations for \( a_1 \) and \( a_2 \) analogous to \( \mathbf{Eq.}(21) \) but this time \([\ldots]_{cc}\) would mean the proportionality to \( \cos k_1 r \cos qz \) or \( \cos k_2 r \cos qz \), respectively. Since both equations have the same structure, we will discuss that for \( a_1 \) only and, for the sake of brevity, we will mention only the terms containing \( a_2 \), the other terms are the same as for the one-sinusoid case discussed above.

For clarity sake, we repeat \( \mathbf{Eq.}(21) \) with a minor change for the present case:

\[
A_2 a_1 + [B P_z^3 + 2 q_{11} P_z u_{zz} + 2 q_{12} P_z (u_{xx} + u_{yy})]_{cc} = 0. \tag{96}\]

It is straightforward to find that the \( a_2 \)-containing term stemming from \([P_z^3]_{cc}\) is \( 9 a_1 a_2^2 / 8 \). From \( \mathbf{Eq.}(26) \), one sees that \([P_z u_{zz}]_{cc} = a_1 \left(u_{zz}^{(0)} + \frac{1}{2} u_{zz}^{(1)}\right)\), recall that now we consider the case \( p = 0 \), and the contribution of this term is

\[
-\frac{3 q_{11}}{8 \lambda_1} a_1 a_2^2. \]

Now

\[
[P_z (u_{xx} + u_{yy})]_{cc} = a_1 \left( u_{1,xx}^{(3)} + u_{1,yy}^{(3)} \right) + \frac{a_1}{4} \left( u_{1,xx}^{(4)} + u_{1,yy}^{(4)} \right) + \frac{3}{4} a_2 (u_{xx}^+ + u_{yy}^+ + u_{xx}^- + u_{yy}^-)
\]

and contribution of this term to the equation of state is

\[
-q_{12} \frac{3 a_1 a_2}{8} \left[f(\theta^+ + f(\theta^-)\right].
\]

Finally, the constituent equation for \( a_1 \) takes the form:

\[
A_2 a_1 + \frac{9}{16} a_1^3 B_2 (\theta_1) + \frac{9}{8} a_1 a_2^2 B_3 (\theta^+, \theta^-) = 0,
\]

where we have introduced

\[
B_3 (\theta^+, \theta^-) = B - \frac{2 q_{11}}{3 \lambda_1} - \frac{2}{3 q_{12}} \left[f(\theta^+) + f(\theta^-)\right]. \tag{97}\]
Similarly to the case of one sinusoid, we recover the free energy:

\[ F = \frac{A_2}{8}(a_1^2 + a_2^2) + \frac{9}{256} B_2(\theta_1) a_1^4 + \frac{9}{256} B_2(\theta_2) a_2^4 + \frac{9}{64} B_3(\theta^+, \theta^-) a_1^2 a_2^2, \]  

(98)

Recall that the square symmetry suggests that \( B_2(\theta_1) = B_2(\theta_2) \), and \( B_2(\theta) \) is given by Eq. (57) and (97).

Turning to examining the sign of the discriminant \( Z \), we should demonstrate that the positiveness of \( B \) allows for the checkerboard domain structures and the limits of absolute instability of a single domain state in thin films of cubic ferroelectric films on a misfit substrate. On the compressive substrate, the cubic ferroelectric

\[ B_2 - 2B_3 = -B + \frac{4}{3} q_{12}^2 \left[ f(\theta^+) + f(\theta^-) - \frac{1}{2} f(\theta_1) \right]. \]

One sees that the maxima of \( Z \) correspond to maxima of \( f(\theta^+) \) and \( f(\theta^-) \) [note that \( f(\theta_{\text{max}}^+) = f(\theta_{\text{max}}^-) \) because of the cubic symmetry], which are, automatically, the minima of \( f(\theta_1) \) as we have seen in Sec. VIII. Then,

\[ [B_2 - 2B_3]_{\text{max}} = -B + \frac{4}{3} q_{12}^2 \left[ 2f(\theta_{\text{max}}) - \frac{1}{2} f(\theta_{\text{min}}) \right]. \]

Using the values of \( f(\theta_{\text{max}}) \) and \( f(\theta_{\text{min}}) \) found in Sec. VIII we find that if \( \lambda_2 + 2\mu > \lambda_1 \),

\[ [B_2 - 2B_3]_{\text{max}} = -B + \frac{4}{3} q_{12}^2 \left( \frac{2}{\lambda_1} - \frac{1}{\lambda_1 + \lambda_2 + 2\mu} \right) = -B + \frac{4}{3} q_{12}^2 \left( \frac{2(\lambda_2 + 2\mu) + \lambda_1}{\lambda_1(\lambda_1 + \lambda_2 + 2\mu)} \right) \]

(99)

and if \( \lambda_2 + 2\mu < \lambda_1 \),

\[ [B_2 - 2B_3]_{\text{max}} = -B + \frac{4}{3} q_{12}^2 \left( \frac{4}{\lambda_1 + \lambda_2 + 2\mu} - \frac{1}{2\lambda_1} \right) = -B + \frac{4}{3} q_{12}^2 \left( \frac{7\lambda_1 - \lambda_2 - 2\mu}{2\lambda_1(\lambda_1 + \lambda_2 + 2\mu)} \right) \]

(100)

To prove that the checkerboard structure can be stable, at least in principle, with respect to small fluctuations, we should demonstrate that the positiveness of \( [B_2 - 2B_3]_{\text{max}} \) is compatible with the conditions \( B_2 > 0 \) and \( [B_2 - 2B_3]_{\text{min}} > 0 \) which guarantee the global stability of the system. We do not intend to perform an exhaustive analysis but want only to demonstrate that this is possible under certain conditions, unlike in the case without the elastic coupling. As an example, we consider a system with a weak elastic anisotropy which is valid for the perovskites, i.e. we shall assume \( \lambda_2 + 2\mu \simeq \lambda_1 \), and \( q_{11} = 0 \). Both Eq. (99) and Eq. (100) then give

\[ [B_2 - 2B_3]_{\text{max}} \simeq -B + \frac{2q_{12}^2}{\lambda_1} \]

(101)

and give for the positiveness of \( [B_2 - 2B_3]_{\text{max}} \) the same condition: \( q_{12}^2 > 3B\lambda_1/2 \) while the condition \( B_2 > 0 \) now reads \( q_{12}^2 < 3B\lambda_1/2 \). One sees that for a nearly elastically isotropic ferroelectric with \( q_{11} = 0 \), the checkerboard structure is at least metastable if

\[ 3B\lambda_1/2 > q_{12}^2 > B\lambda_1/2 \]

Of course, the above set of the material coefficients looks fairly exotic but it is just an example aimed at nothing more but demonstration that the checkerboard domain structures are permitted due to the elastic coupling when certain conditions on the material coefficients are met.

In case of real perovskite films the checkerboard structure is not stable. To see this, we can rewrite Eq. (101) in the form:

\[ [B_2 - 2B_3]_{\text{max}} \simeq -\bar{B} - \frac{2}{\lambda_1} (q_{11}^2 - q_{12}^2) < 0. \]

Indeed, we have already mentioned above that for the perovskites \( q_{11}^2 > q_{12}^2 \). Also, \( \bar{B} > 0 \) there. Therefore, in the perovskites the checkerboard domain structure is absolutely unstable.

VIII. CONCLUSIONS

With the use of the Landau-Ginzburg-Devonshire theory, we have studied the effects of polarization-strain coupling when defining the character of equilibrium domain structures and the limits of absolute instability of a single domain state in thin films of cubic ferroelectric films on a misfit substrate. On the compressive substrate, the cubic ferroelectric
behaves substantially as a uniaxial ferroelectric with the polar axis perpendicular to the film. The film is sandwiched between the electrodes that do not provide a perfect screening of the depolarizing field because of the finite Thomas-Fermi screening length. Such a system is exemplified by (100) BaTiO$_3$/SrRuO$_3$/SrTiO$_3$ film and similar perovskite structures. Quantitative results have been obtained for BaTiO$_3$, PbTiO$_3$, and Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$. We have found that close to the paraelectric-ferroelectric phase transition or at the film thicknesses close to the minimal thickness compatible with the ferroelectricity, the equilibrium domain structure in perovskites is the stripe-wise one with the stripes parallel (perpendicular) to the cubic axes in BaTiO$_3$, PbTiO$_3$, while running at 45° to cubic axes in Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$. The energy of the domain structure depends very weakly on the stripe orientation, the maximum change proves to be well below 1% in all three cases. We found that because of the polarization-strain coupling a competing checkerboard domain structure may, at least in principle, be equilibrium or metastable when certain conditions on the material constants are met, but we are not aware of any material system with such conditions. The limit of absolute instability of the single domain state changes due to the polarization-strain coupling. Thus, the interval where the absolute instability is absent, meaning a metastability in the cases at hand, widens in perovskites in agreement with the earlier conclusion by Pertsev and Kohlstedt. However, this effect is much smaller than that claimed by them. Increase of the metastability range is substantial in BaTiO$_3$ and PbTiO$_3$ where the absolute instability limit becomes close to what is often called the "critical thickness for ferroelectricity" $L^{cD}_{SD}$ Fig.5 but without accounting for the domain formation, which sets in first and prevents the system from ever reaching this point. The effect is much smaller in Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$. We have found also that the polarization-strain coupling can lead to narrowing of the region of relative stability of the single domain state under certain conditions on the material constants, but we are not aware of an experimental realization of these conditions.

AIP has been partially supported by Ministry of Science and Education of Russian Federation (State Contract # 02.740.11.5156).

1. D.J. Kim, J.Y. Jo, Y.S.Kim, Y.J. Chang, J.S. Lee, J.-G. Yoon, T.K. Song, and T.W. Noh, Phys. Rev. Lett. 95, 237602 (2005).
2. Y.S.Kim, D.H.Kim, J.D.Kim, Y.J.Chang, T.W.Noh, J.H.Kong, K.Char, Y.D.Park, S.D.Bu, J.-G.Yoon, J.-S.Chung, Appl. Phys. Lett. 86, 102907 (2005).
3. Y.S. Kim, J.Y. Jo, D.J. Kim, Y.J. Chang, J.H. Lee, and T.W. Noh, T.K. Song, J.-G. Yoon, J.-S. Chung, S. I. Baik and Y.-W. Kim, C.U. Jung, Appl. Phys. Lett. 88, 072909 (2006).
4. J. Junquera and P. Ghosez, Nature 422, 506 (2003).
5. A.M. Bratkovsky and A.P. Levanyuk, Appl. Phys. Lett. 89, 253108 (2006).
6. A.M. Bratkovsky and A.P. Levanyuk, J. Comput. Theor. Nanosci. 6, 465 (2009); arXiv:0801.1669v4 [cond-mat.mtrl-sci].
7. P.Aguado-Puente and J.Junquera, Phys. Rev. Lett. 100, 177601 (2008).
8. E.V. Chensky and V.V. Tarasenko, Sov. Phys. JETP 56, 618 (1982) [Zh. Eksp. Teor. Fiz. 83, 1089 (1982)].
9. N.A. Pertsev and H. Kohlstedt, cond-mat/0603762.
10. N.A. Pertsev and H. Kohlstedt, Phys. Rev. Lett. 98, 257603 (2007).
11. N.A. Pertsev and H. Kohlstedt, Phys. Rev. Lett. 100, 149702 (2008).
12. A.M. Bratkovsky and A.P. Levanyuk, Phys. Rev. Lett. 100, 149701 (2008).
13. A.M. Bratkovsky and A.P. Levanyuk, Phil. Mag. 90, 113 (2010).
14. A.K. Tagantsev and G. Gerra, J. Appl. Phys. 100, 051607 (2006).
15. N.A. Pertsev, A.G. Zemlibotov, and A.K.Tagantsev, Phys. Rev. Lett. 80, 1988 (1998).
16. Y.Yu, S.Y. Hu, Z.K. Liu, and J. Chen, Acta Mater. 50, 395 (2002).
17. G. Sheng, J.X. Zhang, Y.L. Li, S. Choudhury, Q.X. Jia, Z.K. Liu, and J. Chen, J. Appl. Phys. 104, 054105 (2008).
18. J. Hlinka, Ferroelectrics 375, 132 (2008); J. Hlinka and P. Mártón, Phys. Rev. B 74, 104104 (2006); P. Marton, I. Rychetsky, and J. Hlinka, Phys. Rev. B 81, 144125 (2010).
19. V. Dvorak and V. Janovec, Jpn. J. Appl. Phys. 4, 400 (1965).
20. J.Fousek and M.Safrankova, Jpn. J. Appl. Phys. 4, 403 (1965).

In cubic crystals the usual notations for the elastic constants are $\lambda_1 = c_{11}$, $\lambda_2 = c_{12}$, $\mu = c_{44}$. We have used the following values of the parameters in the SI units:
(i) for BaTiO$_3$: $c_{11}$ = 1.755 $\times 10^{11}$Nm$^{-2}$, $c_{12}$ = 8.464 $\times 10^{10}$Nm$^{-2}$, $c_{44}$ = 1.082 $\times 10^{11}$Nm$^{-2}$, $q_{11} = 1.203 \times 10^{10}$Jm$^{-2}$, $q_{12} = -1.878 \times 10^{9}$Jm$^{-2}$, $B = 3.6 \times 10^{8}$ Jm$^{-3}$C$^{-4}$, $G = 0.3A^2$ (Ref. 18);
(ii) for PbTiO$_3$: $c_{11}$ = 1.746 $\times 10^{11}$Nm$^{-2}$, $c_{12}$ = 7.937 $\times 10^{10}$Nm$^{-2}$, $c_{44}$ = 1.111 $\times 10^{11}$Nm$^{-2}$, $q_{11} = 1.141 \times 10^{10}$Jm$^{-2}$, $q_{12} = 4.607 \times 10^{8}$ Jm$^{-2}$, $B = 2.0 \times 10^{8}$ Jm$^{-3}$C$^{-4}$;
(iii) for Pb(Zr$_{0.5}$Ti$_{0.5}$)O$_3$, $c_{11}$ = 1.545 $\times 10^{11}$Nm$^{-2}$, $c_{12}$ = 8.405 $\times 10^{10}$Nm$^{-2}$, $c_{44}$ = 3.484 $\times 10^{10}$Nm$^{-2}$, $q_{11} = 7.189 \times 10^{9}$Jm$^{-2}$, $q_{12} = -2.853 \times 10^{9}$Jm$^{-2}$, $B = 1.43 \times 10^{9}$ Jm$^{-3}$C$^{-4}$.