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Longitudinal and Transversal Polarization Switching in Strained Ferroelectrics

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Using the multicomponent Landau-Devonshire functional we study the field-induced polarization switching in c-oriented ferroelectric phase of epitaxial perovskite film and found that two different regimes are possible: (i) longitudinal switching with change of the polarization modulus and (ii) transversal switching with virtual admixing of the a-component of polarization. The results are specified for the case of barium titanate oxide BaTiO₃ where the crossover line between two types of switching is found at strain-temperature phase diagram.

Keywords Ferroelectric thin films; strain; switching

Introduction

Understanding of mechanism of polarization switching in ferroelectric materials provides an important tool for application and optimization of memory-storage devices. Such study is especially important for technological pseudo-cubic perovskite materials, like e.g. barium titanate oxide BaTiO₃ (BTO) that, depending on polarization orientation, can demonstrate phases with different crystal structure: tetragonal orthorhombic or rhombohedral.

Using the Landau-Devonshire (LD) functional extended up to the 4th order over the multicomponent ferroelectric order parameter $\mathbf{P} = (P_1, P_2, P_3)$, Iwata and Ishibashi [1] theoretically studied the stability of ferroelectric phase under the oppositely applied electric field. They determined the critical field and demonstrated that, depending on the strength of the anisotropic coupling with the crystal lattice, one of the two possible switching mechanisms realizes: either, for strong anisotropy coupling, the switching occurs like in uniaxial one-component ferroelectric by change of the modulus of the longitudinal component P_3 of polarization vector or, if anisotropy is weak, the transversal components P_1 or P_2 virtually admix to P_3 during the switching and polarization effectively rotates as shown in Fig. 1. Such scenario can, for instance, be realized in PbZr_xTi_{1-x}O₃ compounds when x approaches to morphotropic point $x \simeq 0.44$ from above and anisotropy vanishes.

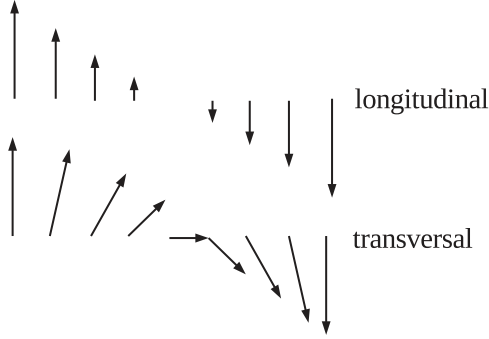


Figure 1. Polarization vector variation during the switching process.

Method

It is well known that the substrate-induced misfit strain acting on thin ferroelectric film can substantially influence the ferroelectric phase transition, change the critical temperature and induce the novel structural phases with respect to the bulk. Notably, the question is actual in the pseudo-cubic perovskite-type ferroelectrics where the applied strain induces the uniaxial anisotropy between the in-plane a - b and out-of-plane c directions.

The classical example of such systems is the pseudo-cubic ferroelectrics BaTiO₃, having the rich strain-temperature phase diagrams as proposed by Pertsev *et al.* [2, 3]. At least five structural phases can be located there: the paraelectric phase with $\mathbf{P} = (0, 0, 0)$ at high temperatures, the c phase with $\mathbf{P} = (0, 0, P_3)$ at high compressive strains, the aa phase with $\mathbf{P} = (P_1, P_1, 0)$ at high tensile strains and either ac phase with $\mathbf{P} = (P_1, 0, P_3)$ or r phase with $\mathbf{P} = (P_1, P_1, P_3)$ at low temperatures and low strains. These phases are separated by the continuous or discontinuous transition lines.

In present article we generalize the results of ref. [1] for the strained epitaxial films and study the dynamics of polarization switching in case when polarization is initially oriented along the perpendicular to substrate c -axis and electric field is opposite to the polarization direction. Starting from the general case of the fourth-order LD model we generalize the results to the particular case of strain-temperature phase diagram of BTO [2, 3], for which an account of the six-order terms is known to be necessary [4, 5, 6].

We begin with the 4th-order anisotropic LD model:

$$\begin{aligned}
 F(P, E) = & \alpha_1 (P_1^2 + P_2^2) + \alpha_3 P_3^2 + a_{11} (P_1^4 + P_2^4) + \alpha_{33} P_3^4 + \alpha_{12} P_1^2 P_2^2 \\
 & + \alpha_{13} (P_1^2 P_3^2 + P_2^2 P_3^2) - E P_3,
 \end{aligned} \tag{1}$$

that can be solved analytically. Coefficients in (1) are, in general, stress- and temperature dependent and $\alpha_{1,3} = A_{1,3}(T - T_{c1,3})$. The electrical field E is applied against polarization that means $E < 0$. The key-difference with respect to the Iwata *et al* [1] case is the anisotropic splitting of critical temperature at P^2 -terms and the possibility to modify the cross-coupling P^4 -terms by strain.

To find the critical field E_c we note that at smaller fields with $|E| < E_c$ the vector of polarization is still oriented along c : $\mathbf{P}(\mathbf{E}) = (0, 0, P_3(E))$ and the field dependence $P_3(E)$

is given by the one-component variational equation:

$$(\partial F/\partial P_3)P_{1,2} = 0 = 2\alpha_3 P_3 + 4\alpha_{33}P_3^3 - E = 0 \quad (2)$$

with $P_3(0) = P_s = (-\alpha_3/2\alpha_{33})^{1/2}$.

By starting from $\mathbf{P}(0) = (0, 0, P_s)$, the application of an opposite electric field leads to the decrease of polarization P_3 which stay positive with $P_{1,2} = 0$ until the switching instability field E_c , that can be calculated as loss of the positive definiteness of the Hessian matrix $H = \partial^2 F/\partial P_i \partial P_j$. The central point here is that, depending in which direction H will lose its stability first, several switching scenarios are possible. The competition occurs between *longitudinal* and *transversal* mechanisms, described by polarization instability along or perpendicular to P_3 , that are characterized by corresponding critical fields E_c^l and E_c^t and associated critical polarizations P_c^l and P_c^t . The instability with lowest amplitude of the critical field will win. In this article we shall study the details of realization of the concrete switching process upon physical parameters of the system.

As was briefly outlined above, to understand, how the switching instability occurs, one has to find the field at which either the longitudinal stability condition:

$$L : (\partial^2 F/\partial P_3^2)P_{1,2} = 0 = 2\alpha_3 + 12\alpha_{33}P_3^2 > 0 \quad (3)$$

or the transversal stability condition:

$$T : (\partial^2 F/\partial P_{1,2}^2)P_{1,2} = 0 = 2\alpha_1 + 2\alpha_{13}P_3^2 > 0 \quad (4)$$

will be broken first. Taking into account that the field dependence of polarization in the instability point is still given by condition (2) we easily find the longitudinal and transversal critical field with corresponding critical polarizations:

$$L : E_c^l = 4/3\alpha_3(-\alpha_3/6\alpha_{33})^{1/2}, P_c^l = (-\alpha_3/6\alpha_{33})^{1/2}, \quad (5)$$

$$T : E_c^t = 2\alpha_3(1 - 2(\alpha_1\alpha_{33})/(\alpha_3\alpha_{13}))(-\alpha_1/\alpha_{13})^{1/2}, P_c^t = (-\alpha_1/\alpha_{13})^{1/2}. \quad (6)$$

In order to make easier the further discussion, we normalize the realized switching field E_s on the longitudinal field E_c^l and consider this ratio R as a function of the parameter $A = 6\alpha_1\alpha_{33}/\alpha_3\alpha_{13}$:

$$R(A) = E_s/E_c^l. \quad (7)$$

One has to distinguish two cases: when $A < 1$ the switching occurs via the longitudinal mechanism at $E_s = E_c^l$ with $R(A) = 1$, and when $A > 1$ switching occurs via transversal scenario at $E_s = E_c^t$ with:

$$R(A) = 3/2(1 - A/3)\sqrt{A}. \quad (8)$$

As one can observe from $R(A)$ dependence, depicted in Fig. 2 the tuning of parameter A by strain or temperature can govern the crossover from one mechanism to another. Note also that the transversal pulse of polarization $P_{1,2}$ can appear even during the longitudinal switching, when the longitudinal component P_3 changes its sign by traversing $P_3 = 0$. Condition $(\partial^2 F/\partial P_{1,2}^2)P_{1,2,3} = 0 = \alpha_1 < 0$ that is *weaker* than $A > 1$ is required for this.

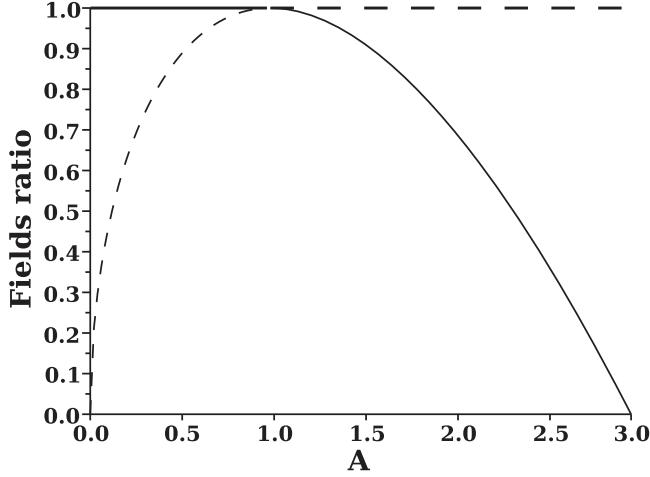


Figure 2. Evolution of the ratio $R = E_s/E_c^l$ as a function of parameter $A = 6\alpha_1\alpha_{33}/\alpha_3\alpha_{13}$ (solid line). The thick solid line and thin solid line respectively correspond to the longitudinal and to the transversal mechanisms. Dashed lines present the analogous ratio, but E_s is replaced by the field of the non-realized switching mechanism: the thick dashed line corresponds to the longitudinal mechanism whereas the thin dashed line to the transversal one.

Switching in Strained BaTiO₃

Now we turn to the practical aspects of realization of the described switching mechanism in case of the epitaxial strained films of BTO for which the six-order terms should also be taken into account. We use the effective LD functional in the proposed by Pertsev et al. form [1]:

$$\begin{aligned}
F(P, E) = & a_1^\star (P_1^2 + P_2^2) + a_3^\star P_3^2 + a_{11}^\star (P_1^4 + P_2^4) + a_{33}^\star P_3^4 \\
& + a_{13}^\star (P_1^2 + P_2^2) P_3^2 + a_{12}^\star P_1^2 P_2^2 + a_{111} (P_1^6 + P_2^6 + P_3^6) \\
& + a_{112} [P_1^4 (P_2^2 + P_3^2) + P_3^4 (P_1^2 + P_2^2) + P_2^4 (P_1^2 + P_3^2)] \\
& + a_{123} P_1^2 P_2^2 P_3^2 - EP_3,
\end{aligned} \tag{9}$$

the values of the coefficients and their dependence on strain being gathered in ref. [2].

Using the same method as above we obtain the variational equation for $P_3(E)$:

$$(\partial F/\partial P_3)P_{1,2} = 0 = 2a_3^\star P_3 + 4a_{33}^\star P_3^3 + 6a_{111} P_3^5 - E = 0 \tag{10}$$

instead of (2) and the longitudinal and transversal stability conditions:

$$L : (\partial^2 F/\partial P_3^2)P_{1,2} = 0 = 2a_3^\star + 12a_{33}^\star P_3^2 + 30a_{111} P_3^4 > 0, \tag{11}$$

$$T : (\partial^2 F/\partial P_{1,2}^2)P_{1,2} = 0 = 2a_1^\star + 2a_{13}^\star P_3^2 + 2a_{112} P_3^4 > 0 \tag{12}$$

instead of (3) and (4).

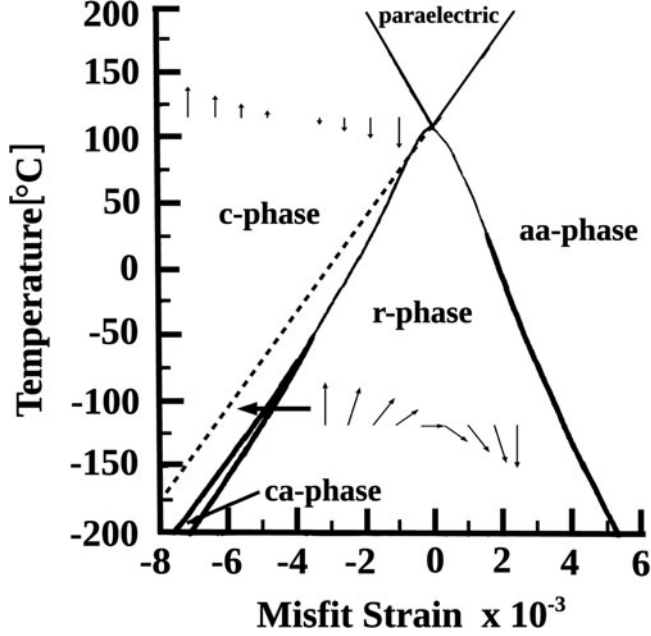


Figure 3. Location of the region of longitudinal and transversal switching mechanism on the temperature-strain phase diagram of substrate-deposited BTO, proposed in ref. [3].

Then we can calculate the longitudinal P_c^l and transversal P_c^t polarizations at the instability fields E_c^t and E_c^l :

$$L : (P_c^l)^2 = ((9a_{33}^{\star 2} - 15a_3^{\star} a_{111})^{1/2} - 3a_{33}^{\star}) / 15a_{111}, \quad (13)$$

$$T : (P_c^t)^2 = ((a_{13}^{\star 2} - 4a_1^{\star} a_{112})^{1/2} - a_{13}^{\star}) / 2a_{112}, \quad (14)$$

but it is not possible to find the analytic expression for E_c^t and E_c^l in simple form. We can however present the condition of crossover from longitudinal to transversal regime when $E_c^t = E_c^l$ just by equating P_c^t to P_c^l :

$$(15a_1^{\star} a_{111} - a_3^{\star} a_{112}) / (15a_{13}^{\star} a_{111} - 6a_{33}^{\star} a_{112}) = (6a_1^{\star} a_{33}^{\star} - a_3^{\star} a_{13}^{\star}) / (15a_1^{\star} a_{111} - a_3^{\star} a_{112}). \quad (15)$$

The corresponding dashed line is located in the Pertsev's et al. strain-temperature phase diagram of BTO [3] as shown in Fig. 3. Note that it is placed very close to the line $a_1^{\star} = 0$, defined as the prolongation of the dashed line of transition from paraelectric to *aa* phase into the nonlinear region of *c*-phase. Above the dashed line the switching mechanism is longitudinal whereas below this line it is transversal.

We have also investigated the influence of the misfit strain on the switching field at room temperature that we obtained after substitution of the defined by (13) and (14) critical polarizations in Eq. 10. Fig. 4 presents the crossover between two regimes which is observed for $S_m = S_{m0} \approx -0.0026$. When $S_m < S_{m0}$ the switching is induced by the longitudinal instability at $E_s = E_c^l$ whereas when $S_m > S_{m0}$ the transversal instability occurs at $E_s = E_c^t$. At S_{m0} , $E_c^l = E_c^t = E_{c0}$. This value has been used to define the field

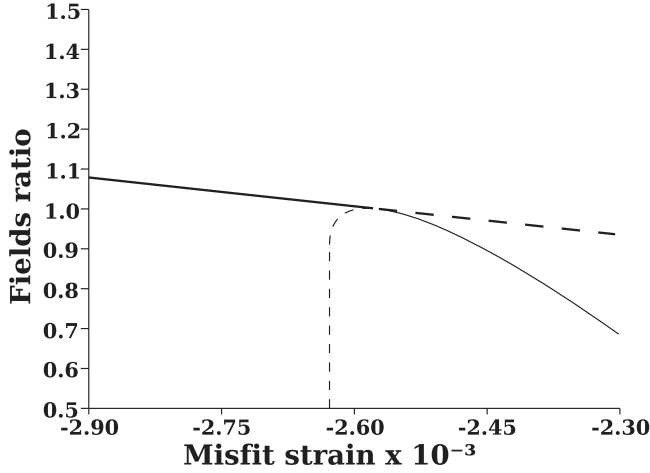


Figure 4. Evolution of the field ratio $R' = E_s(S_m)/E_{c0}$ as a function of the misfit strain S_m (solid line), $E_{c0} = E_c^l(S_{m0}) = E_c^t(S_{m0})$ and $S_{m0} \approx -0.0026$. The thick solid line and thin solid line respectively correspond to the longitudinal and to the transversal mechanisms. With dashed lines, we have represented the evolution of a field ratio, analog to R' except that $E_s(S_m)$ is replaced by the field of the nonrealized switching mechanism at S_m . The thick dashed line and thin dashed line respectively correspond to the longitudinal and to the transversal mechanisms.

ratio $R' = E_s(S_m)/E_{c0}$. One observe that variation of switching field as a function of the misfit strain is more important when switching occurs by the transversal mechanism.

Conclusion

In this article we have demonstrated the existence of two different switching regimes in strained pseudo-cubic perovskite ferroelectric materials in application to BTO film. Depending on the temperature and on the misfit strain one can observe the switching, governed either by the longitudinal instability or by the transversal instability.

The virtual appearance of transversal polarization during transversal switching can be detected by such methods as time-resolved piezo-force microscopy and by the in-field Raman spectroscopy, sensitive to the polarization vector variation. The intriguing possibility would be the observation of the in-plane pulse of transversal pyroelectric current during the polarization switching. Such pulse can serve as the reading indicator of the up-down state modification in the ferroelectric memory cell.

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