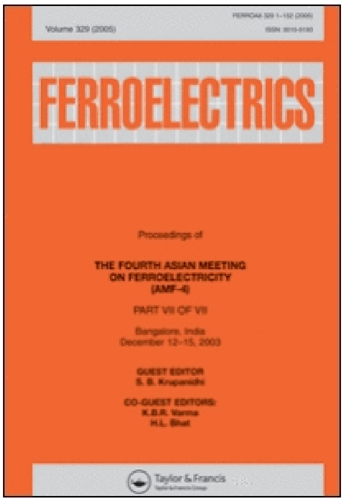

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Temperature Evolution of 180° Ferroelectric Domains in Thin Ferroelectric Films

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Distribution of polarization **P***(x,z) in 180° ferroelectric domains in uniaxial ferroelectric film is studied as function of film thickness* $2a_f$ *and temperature T. It is shown that close to transition temperature and for the very thin films <100nm the polarization has gradual (soft) sin-like profile whereas in thick films and at low temperatures polarization has the flat Kittel-like profile with the sharp domain walls between domains. A simple interpolation formula for polarization profile that recovers both regimes is proposed.*

Keywords Ferroelectrics; thin films; domains

Periodic texture of 180° ferroelectric domains is known to appear in thin ferroelectric film to minimize the energy of depolarization field. In simple Kittel approach [1–3], this domain texture is considered as a set of up- and down- oriented domains, having a flat polarization profile $P(x, z) = \pm P_0$ (hard domains). The domain walls are assumed to be infinitively thin and the boundary effects on the ferroelectric-paraelectric (or ferroelectric-vacuum) interface are neglected. This structure is assumed to be temperature independent and is valid for any width $2a_f$ of the ferroelectric film.

In reality, however, the polarization distribution P(x,z) is more smooth (soft domains), and deviation from Kittel approximation becomes especially important for thin films and high enough temperatures in vicinity of the transition temperature T_c that itself depends on the film thickness, as shown in Figure 1. Note that for very thick films the mono-domain state becomes more preferable that is explained by the alternative mechanism of the depolarization field screening, caused by intrinsic semiconducting charge carriers. In the opposite limit of the very thin films the domain width (scaled according to Kittel as $(2a_f)^{1/2}$ should become of the atomic size).

The objective of the present communication is to study in detail the evolution of polarization profile P(x,z) in the periodic domains structure as function of the temperature and of the film width and to propose the simple interpolation formula that can recover all the regimes of the domain structure, from high temperatures and thin films to low temperatures and thick films. At the same time we concentrate ourselves in the intermediate-width film

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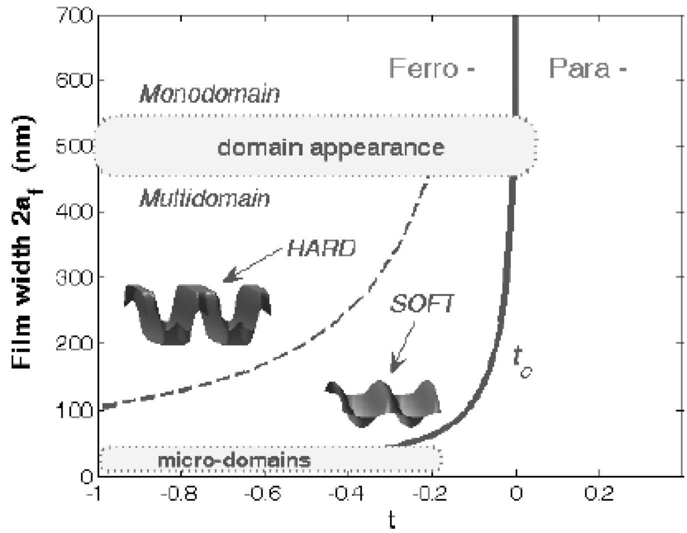

Figure 1. Phase diagram of the domain states in uniaxial ferroelectric as function of the film thickness $2a_f$ and reduced critical temperature $t = 1 - T/T_0$ where T_0 is the bulk critical temperature. In the polydomain region the phase diagram scales as $1/|t|$, as is explained in the text. Profiles of Hard and Soft domains were obtained by numerical solution of equations (4), (5) and (6) using the finite-element PDE toolbox of FemLab [6]. (See Color Plate I)

region (somewhere between 500 and 20nm) in which polydomain state does exit but the domain size is larger then the interatomic distance.

The most appropriate tool to model the polarization distribution is the set of equations constructed from Ginzburg-Landau equations coupled with electrostatic equations describing the depolarization field. These equations, firstly derived by Chensky and Tarasenko [4] (see also [5]), are obtained by variation of the general energy functional:

$$F = \int \phi(P, E)dxdz, \quad \phi(P, E) = \phi(P, 0) - EP - \frac{1}{8\pi}E^2 \tag{1}$$

where $\mathbf{E} = (E_x, E_z)$, $\mathbf{P} = (P_x, P_z)$ and the field-independent part

$$\Phi(P, 0) = \frac{4\pi}{\varepsilon_\perp}\frac{1}{2}P_x^2 + \frac{4\pi}{\varepsilon_{i//}}\frac{1}{2}P_{zi}^2 + f(P) \tag{2}$$

includes the transversal, and non-polar longitudinal noncritical contributions ($\varepsilon_\perp, \varepsilon_{i//} \gg 1$). The nonlinear Ginzburg-Landau energy for the order parameter depends on the spontaneous z-oriented polarization P (assuming that $P_z = P_{zi} + P$) and is written as:

$$f(P) = \frac{4\pi}{\chi_{//}}\left[\frac{t}{2}P^2 + \frac{1}{4}P_0^2P^4 + \frac{\xi_{0x}^2}{2}(\partial_x P)^2 + \frac{\xi_{0z}^2}{2}(\partial_z P)^2\right] \tag{3}$$

Variation of (1) by P and electrostatic potential φ ($E = -\nabla\varphi$) and exclusion of the non-critical variables P_x and P_{zi} gives the system of Chensky equations that describes the ferroelectric transition with the self-action of depolarizing field:

$$-\frac{\chi_{//}}{4\pi}\partial_z\varphi = \left[t - \xi_{0x}^2\partial_x^2 - \xi_{0z}^2\partial_z^2\right]P + (P/P_0)^2P$$

$$\left(\varepsilon_{i//}\partial_z^2 + \varepsilon_\perp\partial_x^2\right)\varphi = 4\pi\partial_z P \tag{4}$$

These equations should be completed by Poisson equation for paraelectric media in which ferroelectric film is embedded:

$$\left(\partial_x^2 + \partial_z^2\right)\varphi^{(p)} = 0 \tag{5}$$

and by boundary conditions at the Para-Ferro interface

$$\partial_z \varphi - \varepsilon_p \partial_z \varphi^{(p)} = 4\pi P, \quad \varphi = \varphi^{(p)}, \quad \partial_z P = 0 \tag{6}$$

that are also obtained as result of variation of (1).

Although the complete set of equations (4), (5) and (6) can be solved numerically [6] (two characteristic examples corresponded to the hard- and soft domain profile are given in Fig. 1) it would be more convenient to have the more general analytical approach permitting generalize the numerical results for the overall parameter region.

Consider first the general properties of equations (4), (5) et (6) rewriting them in dimensionless variables z', x', t', P', φ' defined by:

$$z = a_f z', x = \tau^{-1/2} P_0 P', t = \tau t',$$
$$P = \tau^{1/2} P_0 P', \varphi = \frac{1}{\chi_{//}} \tau^{3/2} a_f P_0 \varphi' \tag{7}$$

With

$$\tau = \left(\frac{\chi_{//}}{\varepsilon_\perp}\right)^{1/2} \frac{\xi_{0x}}{a_f}$$

(the rescaled film width becomes now $2a'_f = 2$) and neglecting the small terms $\frac{\varepsilon_\perp}{\chi_{//}} \frac{\xi_{0z}}{a_f} \partial_z^2$ and $\frac{\varepsilon_{0//}}{\chi_{//}} \frac{\xi_{0x}}{a_f} \partial_z^2$.Omitting the prime index we obtain a new equations set that is independent on the material parameters and therefore is universal for any ferroelectric film:

$$\left(t - \partial_x^2\right) P + P^3 = -\frac{1}{4\pi} \partial_z \varphi^{(f)} \tag{8}$$

$$\partial_x^2 \varphi^{(f)} = 4\pi \partial_z P \tag{9}$$

The corresponding generating functional

$$F = \int \left[4\pi \left(\frac{1}{2} t P^2 + \frac{1}{4} P^4 + \frac{1}{2}\left(\partial_x P\right)^2\right) - \frac{1}{8\pi}(\partial_x \varphi)^2 + P \partial_z \varphi\right] dx dz \tag{10}$$

modifies the boundary conditions (6) as:

$$P = 0, \quad \varphi = \varphi^{(p)} \tag{11}$$

Scaling properties (7) lead to several universal relations for physical properties of multidomain ferroelectric films that will be discussed at the end of the article.

Now we shall try to analyze the temperature dependence of the polarization profile P(x, z, t) that is the solution of new parameter-independent equations (8), (9) and (11), presenting it in a variational form of simple x-periodic function

$$P = f(z) sn\left[\frac{4K(m_1)}{2d} x, m_1\right], \quad f(0) = f(2) = 0 \tag{12}$$

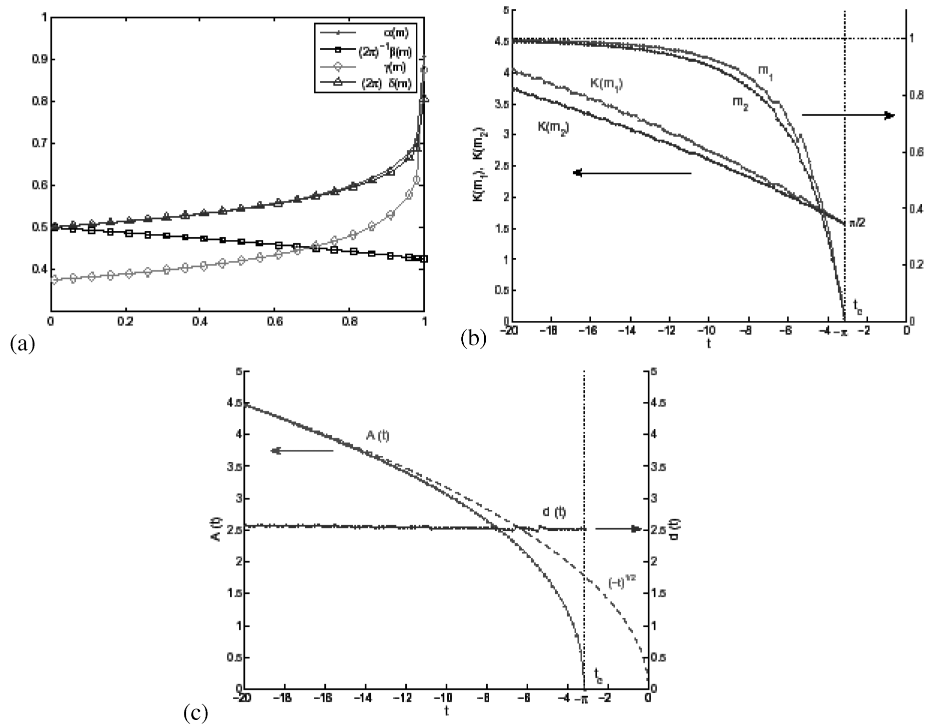

Figure 2. (a) Coefficients $\alpha(m)$, $\beta(m)$, $\gamma(m)$ and $\delta(m)$ that enter in variational functional (13), (b) temperature dependencies of the elliptic arguments m_1 and m_2 and of the elliptic integrals $K(m_1)$ and $K(m_2)$, (c) temperature dependencies of the domain amplitude $A(t)$ and domain lattice period $d(t)$. (See Color Plate II)

that, depending on parameter m_1 ($0 < m_1 < 1$) covers all the domain profiles from the soft ones (at $m_1 \sim 0$) to the hard ones (at $m_1 \sim 1$). Considering m_1, q and function $f(z)$ as variational parameters we substitute (12) into functional (10) and integrate it over domain structure period $2d = q^{-1}4K(m_1)$. After some algebra and nothing that according to Eq. (9) ($\varphi = 4\pi f'(z) \int\int p(x'')dx''dx'$) we present the resulting functional as:

$$F = 4\pi \int \frac{1}{2}\left[\alpha(m_1)t + \frac{4K(m_1)\beta(m_1)}{(2d)^2}\right]f^2 + \frac{1}{4}\gamma(m_1)f^4 + \frac{1}{2}\delta(m_1)(2d)^2 f'^2 dz \quad (13)$$

in which coefficients are expressed via complete elliptic integrals of the first and second kind $K(m)$, $E(m)$ [7] as:

$$\alpha(m) = \langle sn^2(x,m)\rangle = \frac{1}{m}\left[1 - \frac{E(m)}{K(m)}\right]$$

$$\beta(m) = 4K(m)\langle(sn'u)^2\rangle = 4K(m)\frac{1}{3}[2-(1+m)\alpha(m)]$$

$$\gamma(m) = \langle sn^4(x,m)\rangle = \frac{1}{3m}[2(1+m)\alpha(m) - 1]$$

$$\delta(m) = \frac{\langle S^2(x,m)\rangle}{[4K(m)]^2} = \frac{8}{m[4K(m)]^2}\sum_{l=1,3,5}^{\infty}\left[\frac{1}{l}\frac{q^{l/2}(m)}{1-q^l(m)}\right]^2 \quad (14)$$

where $q(m) = \exp[-\frac{K(1-m)}{K(m)}\pi]$, $S(x, m) = \int sn(u, m)du$ and $<\ldots>$ is the average over the period. Dependencies α(m), β(m), γ(m) and δ(m) are presented in Fig. 2a.

Variational Euler-Lagrange minimum of (13) is given by the function:

$$f = A(t, m_1, m_2)sn[K(m_2)z, m_2] \tag{15}$$

with

$$A(t, m_1, m_2) = 2d(t, m_1, m_2)K(m_2)\sqrt{\left(2\frac{\delta(m_1)}{\gamma(m_1)}m_2\right)} \tag{16}$$

that matches the boundary conditions $f(0) = f(2) = 0$ providing that dependence $d(t, m_1, m_2)$ is fixed by biquadratic equation:

$$(2d)^4\delta(m_1)(1 + m_2)K^2(m_2) + (2d)^2\alpha(m_1)t + 4K(m_1)\beta(m_1) = 0 \tag{17}$$

Back substitution of (15) into (13) gives:

$$F = -4\pi\frac{1}{4}\gamma(m_1)\int f^4(z)dz = -\frac{1}{2}\pi\gamma(m_1)A^4(t, m_1, m_2)\gamma(m_2) \tag{18}$$

Collecting now all the results together, we present the final variational solution as:

$$P = A(t, m_1, m_2)sn[K(m_2)z, m_2]sn\left[\frac{4K(m_1)}{2d}x, m_1\right] \tag{19}$$

where the temperature dependencies of m_1 and m_2 and of elliptic integrals $K(m_1)$ and $K(m_2)$ are found by minimization of functional (18) over m_2 and m_1 and are presented in Fig. 2b. The corresponding temperature dependencies of the amplitude A(t) and domain lattice period d(t) are given in Fig. 2c.

Formula (19) that presents the simple approximation for the domain texture at arbitrary temperature and at arbitrary film width is the principal result of the present communication. Several important conclusions can be extracted from dependence (19) and from scaling relations (7):

- Critical temperature of Paraelectric - polydomain Ferroelectric transition (in dimensionless units) is equal to $t_c = -\pi$. In dimensional units it scales as $\tau t_c = -(\frac{\chi_{//}}{\varepsilon_\perp})^{1/2}\frac{\xi_{0x}}{a_f}\pi$.
  All another physical properties of the polydomain state, including the shown in Fig.1 phase diagram scale the same way.
- Domain width is almost temperature independent and approximately equals (in dimensional units) to $2d = \sqrt{-8t_c} = \sqrt{2}(2\pi)$. In dimensional units it scales as $2d = \tau^{-1/2}\xi\sqrt{2}(2\pi)$ in agreement with Kittel low.
- Characteristic scale of the polarization relaxation close to paraelectric - ferroelectric interface is governed by the elliptic integral $K(m_2)$. Its temperature dependence is almost equal to the temperature dependence of the domain wall width, defined by parameter $K(m_1)$.

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References

1. C. Kittel, *Phys. Rev.* **70**, 965 (1946)
2. L. D. Landau and E. M. Lifshitz, Electrodynamics of Continuous Media (Elsevier, New York,1985)
3. A. M. Bratkovsky and A. P. Levanuk, *Phys. Rev. Lett.* **84**, 3177 (2000).
4. E. V. Chensky and V. V. Tarasenko, *Sov. Phys. JETP* **56**, 618 (1982) [Zh. Eksp. Teor. Fiz. **83**, 1089 (1982)]
5. V. A. Stephanovich, I. A. Luk'yanchuk, and M. G. Karkut, *Phys. Pev. Lett.* **94**, 047601 (2005)
6. F. De Guerville, I. Lukyanchuk, L. Lahoche, and M. El Marssi, *Mat. Sci. and Eng.*, **B120**, 16 (2005)
7. M., Abramowitz and I. A. Stegun (eds.) Handbook of mathematical functions (10ed., NBS, 1972).