Elastic domain structure and the transition between polydomain and monodomain states in thin epitaxial films

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We consider an interesting and practically important case of elastic domain structure, which is the analog of the c/a domain pattern with 90° domains in ferroelectric (FE) perovskites and is solvable analytically for arbitrary misfit strain. There is no critical thickness below which the domain structure cannot exist when the ''extrinsic'' misfit is zero and the domains are of equal width. At the boundary of the polydomain-monodomain transition the pattern consists of narrow domains of a minority phase with growing separation between them, which is characteristic of a continuous topological phase transition. The dynamic stiffness of the domain structure diverges while the static stiffness vanishes at this transition. We discuss the implications of the present results for FE films and ceramics.

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Equilibrium domain structures in epitaxial ferroelectric (FE) films may appear even in the case of complete compensation of the depolarizing electric fields by the electrodes or finite conductivity of the film. This takes place if the film is also a ferroelastic. The formation of a ferroelastic domain structure was first considered a while ago as a mechanism to relax a lattice misfit with a substrate.[1] In widely studied perovskite ferroelectrics, which are improper ferroelastics, several elastic domain (and heterophase) structures were predicted.[2–4] One of the typical structures, the so-called $c/a/c/a\ldots$ domain pattern, is of a special fundamental and practical interest. First, the ratio of the widths of the c and a domains changes with external conditions (film thickness and misfit strain). At some parameters the domain pattern does not exist; i.e., there is a phase transition between multi- and monodomain states. Although the corresponding phase diagrams have been discussed by several authors (see Refs. 3 and 4 and references therein), the behavior close to the phase transition was not studied. A numerical study[5] has predicted the response of the c/a domain structure to be large, even giant for some specific values of the (''extrinsic;'' see below) misfit w and the film thickness l, and some data were even interpreted in this way.[6] The enhancement of the response was expected near the border of the existence of the c/a structure in the $w-l$ plane, i.e., near the polydomain-monodomain phase transition. Previous analyses, mainly numerical, have not actually specified the system parameters needed to realize the giant susceptibility. We attempt, therefore, an exact solution of a simplest relevant model, analogous to the actual c/a structures. The present results put strict constraints on the system parameters where the softness of the dielectric and/or piezoelectric response might be large. As a corollary, the existing data[6] cannot be interpreted as being due to a large contribution of the elastic domain walls. We reveal the character of the evolution of the domain structure and show that it is typical of a continuous topological phase transition. The results have important consequences for various applications of FE thin films[7] and ceramics (see Ref. 8 and references therein).

We present here analytical calculations of the energy of a domain pattern analogous to the standard c/a structure. The structure can be viewed, without loss of generality, as a result of a ferroelastic transition in an epitaxial film, which breaks the symmetry of the parent phase and the substrate. All the strains are considered in the reference frame of the high-symmetry phase, and the z axis is selected to be perpendicular to the plane of the film. We consider a system far from the transition, where the pattern consists of domains having a spontaneous strain, $u^0_{xx}-u^0_{zz}$, of opposite signs, separated by domain walls inclined at 45° with respect to the z axis, as dictated by the elastic compatibility conditions and the lattice symmetry, Fig. 1. In ferroelectric perovskites this would correspond to the 90° domains with polarization parallel and perpendicular to the plane. The film consists of a- and c-type domains with widths a_1 and a_2, respectively. From the exact expressions for the free energy F [see Eqs. (6) and (18)], we calculate the period $2a=a_1+a_2$ of the structure, the parameter of asymmetry $\delta=(a_1-a_2)/2a$, and the corresponding stiffness κ. The stiffness κ is closely related to the ''mechanical force constant'' discussed in Ref. 5. We shall consider *two* stiffnesses: the usually measured *dynamic* stiffness κ_∞, calculated at a fixed period of the structure, and the *static* stiffness κ_0, corresponding to the situation where the period of domain structure is allowed to relax via creation or annihilation of the domain walls. The latter corresponds to *very* slow processes, making its observation very challenging. One certainly deals with the dynamic stiffness κ_∞ in applications. We shall show that the two stiffnesses exhibit very different behavior near the transition: the static one diminishes, while the dynamic one diverges. We also study the change of the stiffness with the film thickness and discuss the possibilities of obtaining films with small dynamic stiffness. We consider first the case where the sole origin of the misfit is the spontaneous strain $u^0_{xx}-u^0_{zz}$, appearing at the phase transition. We shall call this misfit ''intrinsic,'' to distinguish it from an ''extrinsic'' misfit of any other origin. The latter includes, e.g., the misfit of the parent phase with the substrate, and other, ''noncritical,'' strain components, which

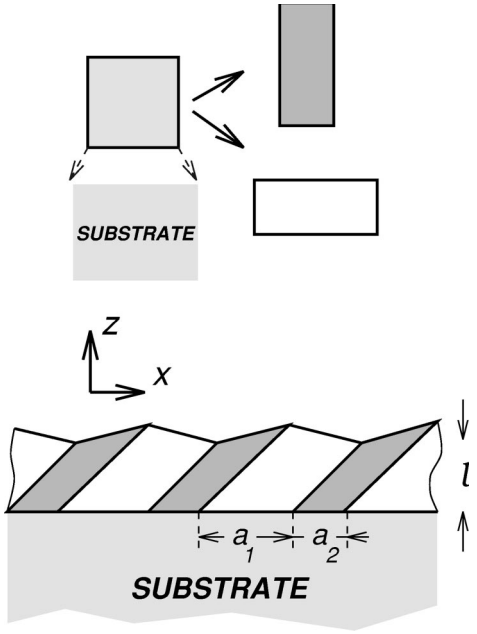

FIG. 1. Schematic of the ferroelastic phase transformation in epitaxial film, when the ''extrinsic'' misfit between the parent phase and the substrate is also present (top panel). As a result, a domain structure with period $2a=a_1+a_2$ is formed in the film of the thickness l (bottom panel).

might appear during the phase transition. The effect of extrinsic strain is a focus of the present paper.

Similar to previous authors[5] we neglect the uncompensated electric fields, which would only *increase* the stiffness. Indeed, we consider an electroded film and find the stiffness of the response in the absence of the external bias voltage, as in Ref. 9. If initially the electric field is absent, i.e., if the sample is in equilibrium, the domain wall shifts would result in the appearance of an additional positive electric energy term in the free energy, $\int dVE^2/8\pi>0$, where E is the depolarizing electric field.[10] Thus, the present consideration gives an estimate for the stiffness from below. It is worth noting also that the intrinsic piezoelectric response is included in effective elastic constants renormalized by the piezoelectric effect. Therefore, it does not matter for the present discussion if the system has an enhanced intrinsic piezoelectric response, e.g., close to a morphotropic transition in $Pb(Zr,Ti)O_3$ transducer (PZT) crystals.

The method of calculating the energy of the domain structure is the same as in Ref. 9 but with more complex equations of state for the stress tensor components σ_{ik}:

$$\sigma_{zz}=(\lambda+2\mu)e_{zz}+\lambda(e_{xx}+e_{yy}), \tag{1}$$

$$\sigma_{xz}=2\mu e_{xz}, \tag{2}$$

with the σ_{xx} and σ_{yy} components obtained from Eq. (1) by cyclic permutation of x,y,z. Here λ,μ are the Lamé coefficients, and $e_{ik}=u_{ik}-u^0_{ik}$ is the elastic strain, with u^0_{ik} the components of the spontaneous strain. In contact with substrate, the film with symmetry-breaking misfit with the substrate must split into domains in such a way that the average in-plane strain will be zero, since the homogeneous strain in the substrate would cost an infinite energy. *Without* the ''extrinsic'' strain the spontaneous strain would alternate from domain to domain (which all would have the same width) as $u^0_{xx}=-u^0_{zz}\equiv\pm u_0$, $u^0_{yy}=0$. On the other hand, *with* the ''extrinsic'' misfit strain w the distribution of the spontaneous strain in the domains would be

$$u^0_{xx}(u^0_{zz})\equiv\pm(\mp)u_0+w, \quad u^0_{yy}=w. \tag{3}$$

We assume for the substrate the same equations of state as Eqs. (1) and (2), but with $u^0_{xx}=u^0_{yy}=u^0_{zz}=0$.

To find the energy of the domain structure, we have to determine the energy of the homogeneous and inhomogeneous (stray) stresses. The energy of the *homogeneous* stresses (F_h), which appears for domains with nonequal widths (i.e., for $\delta\neq0$), can be easily found for an epitaxial film with *any* domain structure, provided that the elastic moduli in all domains are the *same* and the nonlinear effects can be neglected. We can readily find F_h from the general expression for the elastic energy,[9,11]

$$F_{el}=-\frac{1}{2}\int dV\sigma_{ik}u^0_{ik}, \tag{4}$$

as

$$\frac{F_h}{\mathcal{A}}=-\frac{l}{2}(\overline{\sigma}_{xx}\overline{u}^0_{xx}+\overline{\sigma}_{yy}\overline{u}^0_{yy}+\overline{\sigma}_{zz}\overline{u}^0_{zz})$$

$$=Ml[(\overline{u}^0_{xx})^2+(\overline{u}^0_{yy})^2+2\nu\overline{u}^0_{xx}\overline{u}^0_{yy}] \tag{5}$$

$$=Mu_0^2l[(\delta-\delta_0)^2+w^2(1-\nu^2)/u_0^2], \tag{6}$$

where $\delta_0=-w(1+\nu)/u_0$ is the relative extrinsic misfit, $\nu=\lambda/2(\lambda+\mu)$ Poisson's ratio, and $M=2\mu(\lambda+\mu)/(\lambda+2\mu)\equiv E/2(1-\nu^2)$, where E is Young's modulus. The overbar here and below denotes averaging over the film (domain pattern). The condition $\overline{\sigma}_{zz}=0$ was used together with Eq. (1) to obtain for the net planar stresses $\overline{\sigma}_{xx}=2M(\overline{e}_{xx}+\nu\overline{e}_{yy})$ and $\overline{\sigma}_{yy}=\overline{\sigma}_{xx}(x\leftrightarrow y)$, where $\overline{e}_{xx}\equiv\overline{u}_{xx}-\overline{u}^0_{xx}=-\overline{u}^0_{xx}=-u_0\delta-w$ and $\overline{e}_{yy}=-\overline{u}^0_{yy}=-w$, Eq. (3). We have also used the absence of in-plane strains, $\overline{u}_{xx}=\overline{u}_{yy}=0$, imposed by the substrate, and an obvious relation $\overline{u}^0_{xx}=\frac{1}{2}(1+\delta)(u_0+w)+\frac{1}{2}(1-\delta)(-u_0+w)=u_0\delta+w$. Note that Eq. (5) is rather a general expression for the homogeneous strain energy of the epitaxial domain structure and can be applied to other kinds of domain patterns in the epitaxial films (cf. Refs. 9 and 2).

The *stray* energy of inhomogeneous stresses in a stripe-like domain structure periodic in x direction, Fig. 1, is to be found from exact solutions for the strain field produced by the domains. The pattern is defined by the distribution of spontaneous strains $u^0_{xx}(x)$ and $u^0_{zz}(x)$. The condition of local equilibrium, $\partial\sigma_{ik}/\partial x_k=0$, gives two sets of two equations for the displacement components u_x, u_z in the film and the substrate with the use of a standard relation $2u_{ik}=\partial u_i/\partial x_k+\partial u_k/\partial x_i$ and stresses from Eqs. (1) and (2). The boundary conditions are given by the absence of stresses, $\sigma_{zz}=\sigma_{xz}=0$, at the free surface of the film ($z=l$) and at $z\to-\infty$.

Both the strain tensor and the displacement vector are continuous at the film-substrate interface. The original system of partial differential equations is reduced to a system of ordinary differential equations with the use of the Fourier series transform of the functions $u_x(x,z)$ and $u_z(x,z)$:

$$u_{x(z)}=\sum_k u_{x(z)}(k,z)\exp(ikx), \tag{7}$$

where $k=2\pi n/T$, the period of the domain structure $T\equiv 2a=a_1+a_2$, $n=0,\pm 1,\ldots$ (cf. Refs. 12 and 9). We obtain the following equations for the Fourier components of the strain vector in the film $(0<z<l)$:

$$-(\alpha+2)k^2u_x+i(\alpha+1)k\frac{du_z}{dz}+\frac{d^2u_x}{dz^2}=(\alpha+2)\frac{\partial u^0_{xx}}{\partial x}+\alpha\frac{\partial u^0_{zz}}{\partial x}, \tag{8}$$

$$(\alpha+2)\frac{d^2u_z}{dz^2}+i(\alpha+1)k\frac{du_x}{dz}-k^2u_z=(\alpha+2)\frac{\partial u^0_{zz}}{\partial z}+\alpha\frac{\partial u^0_{xx}}{\partial z}, \tag{9}$$

where $\alpha=\lambda/\mu$, and

$$\frac{\partial u^0_{xx}}{\partial x}=-\frac{\partial u^0_{zz}}{\partial x}=\frac{\partial u^0_{xx}}{\partial z}=-\frac{\partial u^0_{zz}}{\partial z}=kR_ke^{ikz}, \tag{10}$$

$$R_k=\frac{2u_0}{kT}(1-e^{-ika_1}). \tag{11}$$

The general solution of this system is given by (cf. Ref. 12)

$$u_x=C_1e^{kz}+C_2e^{-kz}+C_3ze^{kz}+C_4ze^{-kz}-\frac{R_k}{k}e^{ikz}, \tag{12}$$

$$u_z=-iC_1e^{kz}+iC_2e^{-kz}-iC_3\left(z-\frac{\alpha+3}{\alpha+1}\frac{1}{k}\right)e^{kz}+iC_4\left(z+\frac{\alpha+3}{\alpha+1}\frac{1}{k}\right)e^{-kz}+\frac{R_k}{k}e^{ikz}. \tag{13}$$

In the substrate, $z<0$, the equations are the same as Eqs. (8) and (9) with zero right-hand side since the spontaneous deformations are absent in the substrate:

$$-(\alpha+2)k^2u_x+i(\alpha+1)k\frac{du_z}{dz}+\frac{d^2u_x}{dz^2}=0, \tag{14}$$

$$(\alpha+2)\frac{d^2u_z}{dz^2}+i(\alpha+1)k\frac{du_x}{dz}-k^2u_z=0. \tag{15}$$

The general solution of these equations, remaining finite at $z<0$, is

$$u_x=(D_1+D_2z)e^{|k|z},$$

$$u_z=-i\left[D_1+D_2\left(z-\frac{\alpha+3}{\alpha+1}\frac{1}{k}\right)\right]e^{|k|z}. \tag{16}$$

The coefficients in the expressions for the displacements u_x, u_z are found from the boundary conditions. Note that the above equations are similar to those appearing in the corresponding problem of the loss of stability in the same system with the $u_{xx}-u_{zz}$ order parameter at the phase transition.[12] After finding a solution for displacements, Eqs. (12) and (13), we obtain the following simple expression for the stray energy per area $\mathcal{A}$ from Eq. (4):

$$\frac{F_{stray}}{\mathcal{A}}=\frac{2Mu_0^2a}{\pi^3}\sum_{n=1}^{\infty}[1-(-1)^n\cos\pi n\delta]\times\frac{1-(1+2p^2)e^{-2p}}{n^3}, \tag{17}$$

where $p=\pi nl/a$.[13] The series is calculated with the result

$$\frac{F_{stray}}{\mathcal{A}}=\frac{2}{\pi^3}Mu_0^2a\left[\zeta(3)-\mathrm{Re}\,\mathrm{Li}_3(-e^{i\pi\delta})-\mathrm{Li}_3(e^{-b})+\mathrm{Re}\,\mathrm{Li}_3(-e^{-b+i\pi\delta})+\frac{b^2}{2}\mathrm{Re}\ln\frac{1-e^{-b}}{1+e^{-b+i\pi\delta}}\right], \tag{18}$$

where $\mathrm{Li}_n(z)\equiv\Sigma_{s=1}^{\infty}z^s/s^n=z\Phi(z,n,1)$,[14] $b=2\pi l/a$. For $\delta=0$ this formula gives the total elastic energy of a symmetric domain structure (a pattern of opposite domains of equal widths). We have found earlier a somewhat similar formula for another symmetric domain structure of $a_1|a_2$ type.[9] Let us consider the following general cases.

Zero extrinsic misfit $(w=0)$. In this case the energy of the *homogeneous* elastic field is simply $F_h/\mathcal{A}=Mu_0^2l\delta^2$. The *stray* energy has a simple form for the following two limiting cases. In the standard case of narrow domains $(a\ll l)$ and $\delta\ll 1$,

$$\frac{F_{stray}}{\mathcal{A}}=\frac{2}{\pi^3}Mu_0^2a\left[\frac{7}{4}\zeta(3)-\frac{\pi^2\ln 2}{2}\delta^2+\frac{\pi^4}{96}\delta^4\right]. \tag{19}$$

One has to add the surface energy of the domain walls to find the equilibrium domain width

$$F_{dw}/\mathcal{A}=\sqrt{2}\gamma l/a, \tag{20}$$

where $\gamma\equiv Mu_0^2\Delta/\sqrt{2}$ is the energy of the unit surface of the domain walls and Δ is a characteristic microscopic length.[16] The total free energy $F=F_h+F_{stray}+F_{dw}$ is minimal for symmetric domain structure ($\delta=0$ and $a_1=a_2=a$), with the standard (Kittel) domain width (cf. Ref. 9)

$$a=a_K\equiv\left(\frac{2\pi^3}{7\zeta(3)}l\Delta\right)^{1/2}\sim\sqrt{l\Delta}\ll l. \tag{21}$$

Using this result, we see immediately that the high-frequency response of the domain pattern (at a fixed period $2a$ of the domain structure) is

$$\kappa_\infty = \frac{\partial^2}{\partial \delta^2}\left(\frac{F}{\mathcal{A}}\right) = 2Mu_0^2 l\left(1 - \frac{\ln 2}{\pi}\frac{a}{l} + \frac{\pi a}{8l}\delta^2\right) > 0, \quad (22)$$

so the pattern is stable, although it softens because of the negative contribution to the stray energy from the terms $\propto \delta^2$ in Eq. (19). Note also that κ_∞ increases with δ, and this, as we shall see, is a general result.

If the elastic modulus μ were soft, the domains can become *wide*, $a \gg l$ $(b \ll 1)$. There

$$\frac{F_{stray}}{\mathcal{A}} = Mu_0^2 l\left[1 - \delta^2\left(1 - \frac{\pi l}{a}\right) - \frac{8l}{\pi a}\ln\frac{e^{3/4}a}{4\pi l}\right], \quad (23)$$

and one finds a symmetric structure with the large domain width (cf. 9)

$$a = 4\pi e^{1/4} l \exp(\pi\Delta/8l), \quad (24)$$

which would be $\gg l$ if we had a substrate with $\Delta \gg d_{at}$ (cf. the answer for a ferroelectric capacitor[10]). This is contrary to a previously considered Kittel case with narrow domains $a_K \ll l$. In the case of wide domains the response softens considerably, but remains positive,

$$\kappa_\infty = 2\pi M u_0^2 l\frac{l}{a} > 0. \quad (25)$$

It is unlikely, however, that the softness of the present domain structure is of any practical importance. The problem is that even a small extrinsic misfit in very thin films will push the domain structure to the boundary of its existence,[3,5] where it immediately gets *rigid* for experimentally accessible frequencies of external field, as discussed below.

Nonzero extrinsic misfit $(w \neq 0)$. The energy of homogeneous stresses is given by Eq. (6). To find the domain structure close to the boundary of its stability we need all characteristics in the limit $\delta \to 1$, and $a \gg l$ $(b \ll 1)$, i.e., close to a monodomain state. The stray energy in this limit is

$$\frac{F_{stray}}{\mathcal{A}} = \frac{1}{4\pi}Mu_0^2 a(1-\delta^2)^2\left[\ln\frac{4el}{a(1-\delta^2)} - \frac{\pi l}{a}\right]. \quad (26)$$

Note that Roytburd and Yu[2] and Roytburd[15] have approximated the numerically computed stray energy with the functional dependence $(1-\delta^2)^2$, i.e., without the important logarithmic term in the square brackets. Minimizing the total energy $F_{tot} = F_h + F_{stray} + F_{dw}$ with respect to a, we obtain the equation

$$\frac{(1-\delta^2)^2}{4\pi}\ln\frac{4l}{a(1-\delta^2)} = \frac{l\Delta}{a^2}, \quad (27)$$

which has a solution

$$\frac{a}{l} = \frac{1}{(1-\delta^2)\ln^{1/2}(4l/\pi\Delta)}\sqrt{\frac{8\pi\Delta}{l}}, \quad (28)$$

where we have omitted the terms $\propto \ln\ln(4l/\pi\Delta) \ll \ln(4l/\pi\Delta)$. It shows that the domain period diverges as $1/(1-\delta^2)$ close to the phase boundary with the monodomain state. The total energy then takes the form

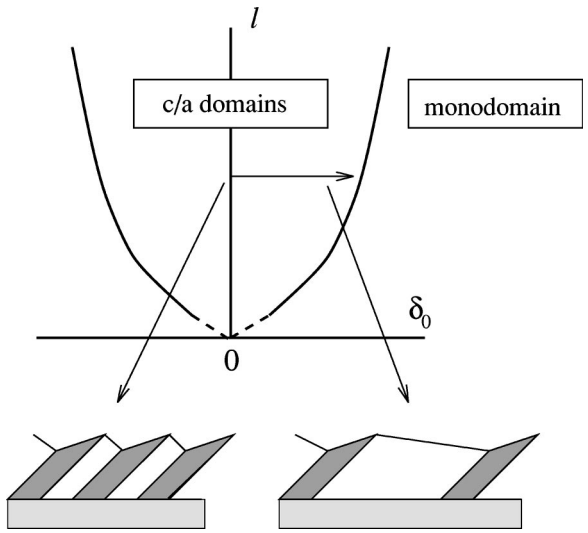

FIG. 2. Schematic of a phase diagram for the present model in coordinates of the film thickness l vs the extrinsic misfit δ_0. The phase boundary is given by Eq. (30), which is valid at almost all thicknesses l. Note that for zero extrinsic misfit $\delta_0 = 0$ the domain structure forms at all thicknesses of the film. With the increase of the extrinsic misfit (horizontal arrow) the domains stretched along the substrate grow to very large sizes, whereas the opposite domains remain narrow (as shown at the bottom of the figure).

$$\frac{F_{tot}}{Mu_0^2 l\mathcal{A}} = (\delta - \delta_0)^2 + \phi(1-\delta^2) - \frac{1}{4}(1-\delta^2)^2, \quad (29)$$

where from the equilibrium the value of δ is to be found (subject to the constraint $\delta^2 \leqslant 1$) with the parameter $\phi \equiv (\mathcal{L}\Delta/2\pi l)^{1/2} \ll 1$, and $\mathcal{L} = \ln(4l/\pi\Delta)$ the logarithm of a large number $\sim l/\Delta \gg 1$.

The transition into a monodomain state $(\delta^2 = 1)$ occurs as a function of the extrinsic relative misfit at $|\delta_0| = 1 - \phi$. This condition suggests a critical thickness of the film, l_c, where the polydomain-monodomain transition takes place at a given misfit δ_0 (i.e., the *phase boundary* in the δ_0-l plane),

$$\frac{l_c}{\Delta} = \frac{1}{\pi(1-\delta_0)^2}\ln\frac{1}{1-\delta_0}. \quad (30)$$

Thus, domain structure may exist only at thicknesses $l > l_c(\delta_0)$. This formula is obtained for $l_c/\Delta \gg 1$, but it also correctly gives the *absence of the critical thickness* of the film when the parameter δ_0 is zero ($l_c = 0$ when $\delta_0 = 0$). Since Δ is very small,[16] the equation for the phase diagram, Eq. (30), is valid almost everywhere. The corresponding phase diagram is shown schematically in Fig. 2.

The absence of a critical film thickness has been suggested earlier for another domain pattern $a_1|a_2$ from numerical computations.[3] This is contrary to the speculations by Roytburd, who obtained $l_c \neq 0$ for $\delta_0 = 0$, apparently as an artifact of the employed approximations.[15] The critical point is approached linearly with δ_0:

$$\delta = 1 - (1 - \phi - \delta_0)/\phi. \quad (31)$$

We see that the slope is $d\delta/d\delta_0 = 1/\phi \gg 1$, so that the approach to the critical point is very steep; it looks almost discontinuous.

We readily obtain the high-frequency (measurable) dynamical stiffness close to the phase boundary,

$$\frac{\kappa_\infty}{Mu_0^2 l} = \frac{1}{1-\delta^2}\sqrt{\frac{8\Delta}{\pi \mathcal{L} l}}\ln\frac{2e^2\mathcal{L}l}{\pi\Delta} \simeq \frac{1}{1-\delta^2}\sqrt{\frac{8\mathcal{L}\Delta}{\pi l}} \to \infty, \tag{32}$$

which *diverges* when $\delta^2 \to 1$, i.e., close to the phase boundary. This is in striking disagreement with the results of Pertsev *et al.*[5] who claimed a softness of the c/a domain structure close to the phase boundary. The static stiffness, which has no practical significance since it requires a very long time to optimize the number of domain walls, is almost constant away from the transition and sharply vanishes with increasing relative misfit δ_0,

$$\frac{\kappa_0}{Mu_0^2 l} \propto 1-\delta^2 \propto 2(1-\phi-\delta_0)/\phi \to 0; \tag{33}$$

see the exact result in Fig. 3. Interestingly, close to the transition into the monodomain state the system splits into two groups of wide (a_1) and narrow (a_2) domains with

$$a_1 = \frac{1}{1-\delta}\sqrt{\frac{8\pi l\Delta}{\mathcal{L}}} \sim \frac{a_K}{(1-\delta)\sqrt{\mathcal{L}}} \to \infty, \tag{34}$$

$$a_2 = \sqrt{\frac{2\pi l\Delta}{\mathcal{L}}} \sim \frac{a_K}{\sqrt{\mathcal{L}}} < a_K. \tag{35}$$

We see that the width of the wide domains diverges and the density of the domain walls decreases, but the width of the narrow domains, a_2, remains small. The narrow domains are somewhat compressed compared to the standard Kittel width a_K, Eq. (21).

It becomes obvious from the present analysis that it is unlikely that one can succeed in making a ''soft'' domain pattern with small κ_∞ in epitaxial thin ferroelectric films with elastic domains. In fact, the only way to do this would be to avoid any extrinsic misfit, i.e., to keep $\delta_0 = 0$, and to reduce the film thickness. However, in this case the interval of the extrinsic misfit strain, which allows the very existence of the domains, is very narrow. Moreover, the proximity to the phase boundary in systems with two kinds of inequivalent domains means that the stiffness rapidly increases even with a slight misfit, which might appear due to, e.g., a minute deviation of the temperature from the value corresponding to zero misfit. We have tacitly assumed above that the stiffness of the domain structure determines its dielectric response. Indeed, one can assume that there is a spontaneous polarization parallel to the film plane in one domain and perpendicular to it in another (as in *a* and *c* domains in perovskites). One can also assume, as in Ref. 5, that $\delta - \delta_{eq} = lP_s E_{ext}/\kappa_\infty$ in an external field, where P_s is the spontaneous polarization, E_{ext} the external electric field, and δ_{eq} is the equilibrium value of δ in zero field. It will be small for large stiffness κ_∞. We have shown earlier[9] that this relation in some cases strongly overestimates the dielectric response. In addition, the neglected contribution of uncompensated electric fields would only make the response stiffer. The conclusion is that the dielectric response of the pattern with inequivalent elastic domains in the epitaxial thin films is actually suppressed. The evolution of this domain structure is typical of a continuous topological phase transition. The present results are likely to apply to bulk ceramic samples, since there the particles are also split into domains, and the response is likely to be affected by the motion of the domain walls in a similar manner.[8]

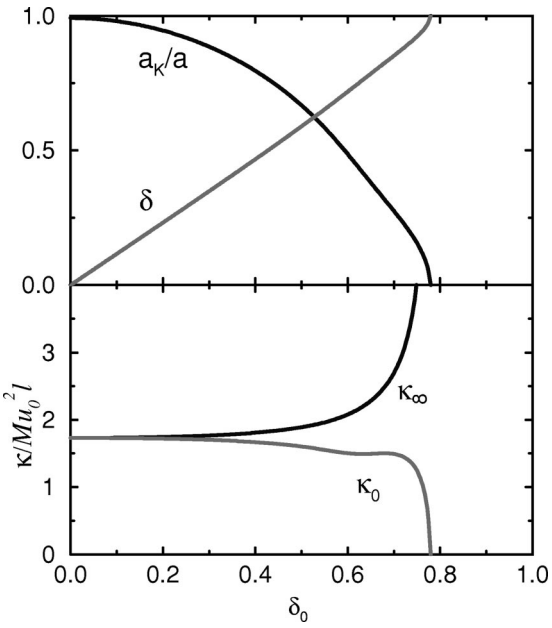

FIG. 3. The relative half-period of the structure a/a_K, where a_K, Eq. (21), is the standard Kittel domain width and the asymmetry parameter $\delta = (a_1 - a_2)/2a$ vs the relative extrinsic misfit δ_0 (top panel). Close to the boundary of existence of the domain structure, where $a \to \infty$, the static stiffness κ_0 vanishes, whereas the (measurable) dynamic stiffness κ_∞ diverges (bottom panel).

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[14] I.S. Gradshteyn and I.M. Ryzhik, *Table of Integrals, Series, and Products*, 5th ed., edited by A. Jeffrey (Academic, New York, 1994), Sec. 9.55.

[15] A.L. Roytburd, J. Appl. Phys. **83**, 228, 239 (1998).

[16] Δ is usually considerably smaller than the atomic length scale d_{at} for proper ferroelastics. For perovskite ferroelectrics, which are improper ferroelastics, it is comparable to the domain wall width. We absorb $\sqrt{2}$, the factor coming from the domain walls being inclined at 45°, into the definition of Δ.