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Thickness-dependent evolutions of domain configuration and size in ferroelectric and ferroelectric-ferroelastic films

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Non-monotonous thickness-dependent ferroelectric and ferroelectric-ferroelastic domain size scaling behaviors were revealed in ferroelectric films, including three distinct regions: (I) a classical $1/2$ power law relationship for thick films, (II) a deviation from the $1/2$ scaling relationship for an intermediate thickness range, and (III) an exponential increase in ultrathin films when decreasing the film thickness. The calculations indicate a much narrower region (II) in ferroelectric films with ferroelectric domains than that with ferroelectric-ferroelastic ones. As the film thickness decreases, the stable domain pattern also changes from a ferroelectric-ferroelastic domain to a ferroelectric one, which leads to the divergence of domain size scaling. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4794005>]

I. INTRODUCTION

Ferroc (ferromagnetic, ferroelectric and ferroelastic) materials have attracted a great deal of attention due to the novel physical properties and potential applications. On cooling below a threshold temperature, i.e., the Curie temperature, domain pattern occurs in ferroic materials. The domain structure and size in ferroics, which depend on various magnetic, electrical, and mechanical boundary conditions, are vital to the applications in data storage devices, actuator, sensors, etc.^{1,2} The stable domain size is determined by the minimum energy condition between the energy of domain wall and the energy of domain itself, which at least include two terms with opposite dependences on the domain size. For thick ferroic films, the domain size is much less than the film thickness (i.e., “dense” domain structure) and the film thickness-dependent size decreases with the classical square root behavior ($1/2$ power law relationship), which has been clarified in ferromagnetic by Landau, Lifshitz and Kittel,^{3,4} ferroelectric by Mitsui and Furuichi,⁵ and ferroelastic materials by Roitburd.⁶ Driven by the miniaturization in ferroic materials, it is indispensable to investigate the domain size scaling behavior in nanoscale systems. The domain size is comparable to the film thickness and the classical $1/2$ power law relationship is not applicable anymore for ferroic thin films less than a definite critical thickness.^{7–13} For example, the square root scaling behavior was reported to break down at critical thickness less than 25 nm for ferromagnetic cobalt films.¹⁴ For thinner ferroic films, the energy from domain itself starts surpassing the domain wall energy. Subsequently, the domain size no longer decreases but increases exponentially as the film thickness decreases.

Although the domain structure and domain size play a crucial role in the properties of ferroic films, a thorough film

thickness-dependent domain size scaling behavior within the published experimental and theoretical data is still under debate. Firstly, the domain size scaling behavior of ultrathin ferroic films remains elusive, particularly for thin films below 100.0 nm. There have been several investigations on the unusual thickness-dependent domain size scaling behavior in ultrathin ferroic films. For example, a recent study showed a striking thickness-dependent domain size scaling behavior with the exponent of 0.59 ± 0.08 in rhombohedral BiFeO₃ (BFO) multiferroic films below 100 nm,¹⁵ in contrast to the classical scaling index of $1/2$. Besides, *anomalous* linear domain size scaling behaviors in tetragonal films were reported with thickness limited to several ten nanometers.^{10,16,17} Secondly, despite the widespread attention in ferroelectric films, the systematic comparison analyses of the domain size scaling behaviors between the ferroelectric domains and ferroelectric-ferroelastic domains are still largely unclear. In ferroelectric films, 180° domains are purely ferroelectric, whereas 90° domains are ferroelectric and ferroelastic. For the sake of simplicity, the 90° ferroelectric-ferroelastic domain is abbreviated to ferroelastic domain. The $1/2$ power law relationship of domain size scaling of 90° domains were extended by Pompe *et al.* to thick ferroelectric films with $a/c/a/c$ or $a_1/a_2/a_1/a_2$ domain structure.^{8,10} In stark contrast to the experimental observation of the deviation from the $1/2$ power law relationship (the power exponent nearly 1.0) of $a/c/a/c$ ferroelastic domain size in ultrathin ferroelectric-ferroelastic PbTiO₃ (PTO) films below 30.0 nm,¹⁶ the scaling behavior of 180° ferroelectric domain size experimentally still follows the conventional $1/2$ relationship in pure ferroelectric PTO ones, even with the film thickness less than 1.0 nm.¹⁸ Furthermore, despite its strong effect on the equilibrium of domain structure and related properties, the depolarization field is usually neglected in ferroelectric films with ferroelastic domain patterns (i.e., $a/c/a/c$) by assuming an ideal screening of the depolarization field near the films surfaces.^{9–12,16,19} To tackle all these issues, it is essential to investigate and

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distinguish the film thickness-dependent evolutions of domain pattern and domain size scaling between ferroelectric and ferroelastic domains.

II. THEORY AND METHODOLOGY

A. Thickness-dependent domain size scaling behaviors in tetragonal ferroelectric PTO films

Although the charged defects and dislocations may exist in real films and affect the associated domain structure and properties, which are expected to affect the domain nucleation and its transition, for simplicity, here we resort to an ideal case for ferroelectric films epitaxially grown on a matched (001)-oriented substrate without charged defects and dislocations. The stable domain patterns and the relationships between domain size W and film thickness H can be determined based on the mechanical and electrical compatibility conditions. To study the domain size scaling behaviors in ferroelectric films, we start with a general thermodynamical model for 180° ferroelectric and 90° ferroelastic domains of tetragonal PTO films that allows us to predict an exact dependence of any H on W . The total energy of ferroelectric PTO films consisting of neutral 180° domain can be expressed as:⁷

$$F_t^{ele} = \frac{1}{2} \frac{P_0^2 H}{\epsilon_0 \epsilon_c} \left(\frac{8g}{\pi^2 R} \sum_{n=1,2,\dots}^{\infty} \frac{1}{n^3} \sin^2 \left(\frac{1}{2} n \pi \right) \frac{1}{1 + g \coth nR} \right) + \frac{\sigma H}{W}, \quad (1)$$

where $g = \sqrt{\epsilon_a \epsilon_c}$ ($\epsilon_a = \epsilon_c$), $R = \frac{\pi H}{2W} \sqrt{\frac{\epsilon_a}{\epsilon_c}}$, ϵ_a, ϵ_c are the permittivities of the ferroelectric, parallel to the a and c crystallographic axes, respectively. P_0 is the spontaneous polarization. The energy terms in the right side of Eq. (1) represent the electrostatic energy and the domain wall energy, respectively. Meanwhile, the total energy of ferroelastic PTO films consisting of 90° ferroelastic domains can be rewritten as¹⁰

$$F_t^{elas} = \frac{GH}{(1-\nu)} (s_a^2 + 2\nu s_a s_c + s_c^2) + \frac{GH}{(1-\nu)} (s_a - s_c)^2 \times \left[2 \left((1+\nu) \frac{s_a}{s_a - s_c} \right) \phi + \frac{2W}{H} f_c \left(\frac{2W}{H}, \phi \right) \right] + \frac{\sqrt{2}\sigma H}{W}, \quad (2)$$

where G is the shear modulus. ϕ is the volume fraction of c domain in the epitaxial film. f_c is a dimensionless function of ϕ and W/H . s_a and s_c are the misfit strains of a -domain and c -domain, respectively.

Minimizing the total energies of ferroelectric films with the reported parameters,^{16,20,21} the exact film thickness-dependent domain size scaling behaviors of ferroelectric and ferroelastic domains in PTO films (black curves) are plotted in Figs. 1(a) and 1(b), respectively. The results show that both the evolutions of size scaling behavior of 90° and 180° domains in PTO films are non-monotonic with the film thickness. For thick films ($H \gg W$), the film thickness-dependent domain size decreases with the conventional 1/2 power law relationship.^{22,23} We label this range as region (I) as shown

FIG. 1. Thickness-dependent domain size in tetragonal PbTiO_3 films: (a) for 180° ferroelectric domains and (b) for 90° ferroelastic domains; Experimental data (Scatter) extracted from Refs. 16, 18, and 24. Both the critical thicknesses (Black dash lines) of ferroelastic PTO films are larger than these of ferroelectric PTO films.

in Fig. 1. The analytic approximate expressions of the 1/2 relationship can be derived for ferroelectric and ferroelastic domains (blue curves),^{7,10} which correspond to the Mitsui and Furuichi's relationship and Roitburd's relationship, respectively

$$W_{approx}^{ele} = \sqrt{\frac{\pi^3 \epsilon_0 \sigma (1 + \sqrt{\epsilon_a \epsilon_c}) H}{8.4 P_0^2}}, \quad (3)$$

$$W_{approx}^{elas} = \sqrt{\frac{11.3 \pi^3 (1 - \nu) \sigma H}{8.4 G (s_a - s_c)^2}}. \quad (4)$$

However, the domain size scaling behavior deviates from the classical 1/2 power law relationship for films thickness below a critical thickness H_1^{crit} . For ferroelectric films, the critical thickness of the deviation is expressed as⁷

$$H_1^{crit} = \frac{5\pi\sigma\epsilon_0}{P_0^2} \sqrt{\frac{\epsilon_c^2}{\epsilon_a}}. \quad (5)$$

The value of H_1^{crit} is 1.8 nm for ferroelectric PTO films as shown in Fig. 1(a). In analogy to the evolution of thickness dependence of ferroelectric domain size scaling behavior, the critical thickness for the divergence in ferroelastic domains can be expressed:⁶

$$H_1^{crit} = \frac{100(1-\nu)\sigma}{G(s_a - s_c)^2}. \quad (6)$$

The value of H_1^{crit} is 25.0 nm for ferroelastic PTO films, which is an order larger than that of ferroelectric PTO ones

(1.8 nm). Both the calculated critical thicknesses agree well quantitatively with the experimental results in ferroelectric/ferroelastic PTO,^{16,18} for instance, the observed thickness-dependent domain scaling behavior of 180° ferroelectric PTO films still obeys the 1/2 power law relationship even for the case where film thickness is at 2.0 nm (shown in Fig. 1(a)). Below the critical thickness H_1^{crit} , the domain size scaling behavior continues to decrease with the film thickness, yet, it begins deviating from the classical 1/2 power law relationship. We label this part of the curve as region (II) as plotted in Fig. 1. However, region (II) shortens significantly in ferroelectrics compared to the corresponding one in ferroelastics. It is shown in Fig. 1 that the range of region (II) is only 0.6 nm for PTO films with ferroelectric domain, in comparison to that 12.0 nm in PTO films with ferroelastic domain. This 12-nm region (II) with the thickness range from 13.0 nm to 25.0 nm in PTO films Fig. 1(b) was theoretically reported before to present a nearly linear behavior of ferroelastic domain size scaling for (*a*/*c*/*a*/*c*) PTO films.¹⁰ Recently, another quasi-linear relationship of domain size scaling behavior was experimentally reported for ferroelastic PTO films thickness in the range from 30.0 to 100.0 nm.¹⁶ However, according to our calculated results, the films with thickness in this range (from 30.0 nm to 100.0 nm) still locate at region (I) in Fig. 1(b), where the thickness-dependent domain size scaling obeys the 1/2 power law relationship rather than the nearly linear one in region (II). This discrepancy between Ref. 16 and our interpretation is most likely due to the technical limitations of extracting values of slopes from an experimental plotting within relatively narrow thickness windows.

There is another critical film thickness H_2^{crit} , below which the domain size no longer decreases rather than increases rapidly and the domain structure tends to change from multi-domain to mono-domain. We label this increasing part of curve as region (III) as plotted in Fig. 1(a). For ferroelectric PTO film, H_2^{crit} is 1.2 nm. However, in PTO films with *a*/*c*/*a*/*c* ferroelastic domain structure, the critical thickness H_2^{crit} is 13.0 nm. The differences of the critical thicknesses between ferroelastic and ferroelectric domains in PTO films become apparent as summarized in Table I, more specifically; both critical thicknesses in films with ferroelastic domains are much larger than these in ferroelectric ones, which imply that ferroelectric 180° domain structure is more stable in ultrathin films. This is strongly consistent with the experimentally observed crossover from 90° ferroelectric-ferroelastic domain (*a*/*c*/*a*/*c*) to 180° ferroelectric one (*c*₁/*c*₂/*c*₁/*c*₂) for PTO ultrathin films with thickness below 12.0 nm.¹⁶ In contrast, the experimentally observed 180° PTO ferroelectric domain size still decreases with the film thickness down to 1.0 nm.^{18,24}

TABLE I. Summary of two critical thicknesses for ferroelectric and ferroelastic domains.

Material	Type of domain	H_{crit1} (nm)	H_{crit2} (nm)
PbTiO₃	Ferroelectric	1.8	1.2
	Ferroelastic	25.0	13.0
BiFeO₃	Ferroelectric	15.0	7.5
	Ferroelastic	445.0	220.0

B. Thickness-dependent domain size scaling behavior in rhombohedral BFO films

In addition to typical tetragonal domains of PTO films, we consider the domain size scaling in rhombohedral BFO films. 109° domains of BFO films are not only ferroelastic domains but also ferroelectric ones. The ferroelectric energy of BFO films can be obtained using Eq. (1), while the total energy consisting of elastic and domain wall energies based on elastic theory can be expressed as:^{12,25}

$$F_t^{109} = \frac{G\omega_s^2 W^2}{2\pi} \left(\frac{8H}{W} \tan^{-1} \frac{W}{2H} - \frac{4H}{W} \tan^{-1} \frac{W}{H} + \ln \left(\frac{4H^2 + W^2}{H^2 + W^2} \right) \right) + \frac{G\omega_s^2 H^2}{\pi} \ln \left(\frac{16H^3 \sqrt{H^2 + W^2}}{(4H^2 + W^2)^2} \right) + \frac{G\omega_d^2 H^2}{2\pi(1-\nu)} \ln \left(\frac{4H^2 + W^2}{H^2 + W^2} \right) + \frac{\sigma H}{W}. \quad (7)$$

For rhombohedral BFO films with 109° domains, the mechanical and the electrostatic conditions influence the domain size dramatically. The film thickness-dependent domain size *W* of 109° domain referred to the elastic and ferroelectric theories are shown in Fig. 2, respectively. Based on the pure ferroelectric theory, the two critical thicknesses of BFO films are 14.0 nm and 7.5 nm as shown in Fig. 2(a), respectively. The remarkable differences are shown in Fig. 2 for these two critical thicknesses using elastic theory (480.0 nm and 220.0 nm). Our calculated results for rhombohedral BFO films further imply that the two critical thicknesses in ferroelastic domains are much larger than these of ferroelectric ones.

FIG. 2. Thickness-dependent domain size in 109° rhombohedral BFO films calculated (a) as ferroelectric domain and (b) as ferroelastic one.

FIG. 3. Schematic of possible thickness-dependent evolution of domain pattern of PTO thin films deposited on small tensile strained substrate. With the increase of the fraction of c domain, the domain pattern changes from $a_1/c_1/a_2/c_2$ to $a_1/c_1/c_2/a_2/c_1$, resulting in the increase of domain periodicity at region (II), and finally to c_1/c_2 . Symbols stand for the polarizations directing to in-plane for a_2 domains. The blue arrows denote the polarization direction. The dash red arrows denote the domain periodicity.

III. THICKNESS-DEPENDENT EVOLUTION OF DOMAIN PATTERN

We note that, in contrast to the experimentally observed increase of domain size with thickness from 12.0 nm to 30.0 nm in PTO films (ferroelastic films with 90° domain structure) deposited on DyScO₃ (DSO), our results show a nearly linear decrease at region (II) as shown in Fig. 1(b). The gap is mainly due to the strong depolarization field effect. Ferroelastic PTO films with out-of-plane polarization are ferroelectric. Nevertheless, it was thought that the depolarization field plays a negligible role in PTO films with elastic domain structure.^{10,12,16} The perfect agreement between theoretical results and experiments in region (I) (Fig. 1(b)) is strongly suggestive of a trivial depolarization field effect for thick PTO films with $a/c/a/c$ domain structure.¹⁶ However, as the relative volume fraction of c domain increases in $a/c/a/c$ domain pattern,¹⁶ the depolarization field energy increases dramatically, which can even be much larger than the energies of elastic domains and domain walls in ultrathin PTO films. In order to reduce the depolarization field, it is essential that a self-driven domain pattern transformation from $a_1/c_1/a_2/c_2$ to $a_1/c_1/c_2/a_2/c_2/c_1$ with alternative 180° out-of-plane polarizations could occur in the region (II). As illustrated in Fig. 3, this transformation leads to an increase of the domain periodicity with the decrease of ferroelastic PTO films thickness, which can explain the experimental observation of increase of domain size (see the represented red star points shown in Fig. 1(b)) in ferroelastic PTO films on DyScO₃ (DSO) with thickness from 12.0 to 30.0 nm. Moreover, despite of a negligible role in thick films, the distinct effect of depolarization field leads to the evolution of domain pattern with the emergence of 180° $c_1/c_2/c_1/c_2$ ferroelectric domains, and affects the domain size scaling behaviors in the region (II) of ferroelastic PTO ultrathin films. To compare, in ferroelastic rhombohedral BFO thin films, the thickness-dependent depolarization field is also indispensable and considered for the evolution between 71° and 109° domain patterns and their size scaling behaviors.^{25,26} In fact, it was theoretically demonstrated that the decrease of the thickness-dependent depolarization field of rhombohedral BFO films could lead to a switch of domain structure from

109° domain to 71° one.²⁶ To further decrease the depolarization field, a similar scenario of the occurrence of 180° ferroelectric domains in rhombohedral BFO thin films was observed experimentally by transmission electron microscopy (TEM), which results in an obvious deviation of domain scaling behavior from the $1/2$ power law relationship.²⁷ Here, we suggest that more detailed dark-field TEM studies are necessary to conduct to further testify the possible domain pattern evolution of ferroelastic PTO thin films proposed in Fig. 3 and check the role of 180° domains in the scaling behavior of region (II).

The results presented above are based on simple geometrical arrangements of domain patterns. It should be pointed out that there are many other factors, for instance, crystal defects, electric charges, and misfit dislocations, which can influence the domain patterns and their size scaling behaviors in ferroic films. Furthermore, different domain patterns, for instance, vertices, vortices, fractals or irregular patterns, rather than the stripes, may form due to the reduction of symmetry in various electric and mechanical boundaries,¹ concomitantly, which result in a wide distribution of domain size and the divergence of domain size scaling behavior ($1/2$ power law).²⁷ In addition, we note that the domain configuration and size scaling behavior are also sensitive to the temperature and sample preparation method and heat-treatment history.^{18,28}

IV. CONCLUSIONS

In summary, we investigated the film thickness-dependent evolution of domain pattern and its size scaling in ferroelectric films. The results provide insight into the analogies and differences of domain size scaling behaviors between ferroelectric and ferroelectric-ferroelastic domains. The evolutions of thickness-dependent domain size scaling in both films are a three-step process: (I) the classical $1/2$ power law relationship for thick films, (II) the deviation from the $1/2$ relationship for intermediate film thickness, and (III) an increase exponentially in ultrathin films when decreasing film thickness. Both the critical thicknesses of ferroelectric films with ferroelectric domains are much smaller in contrast with these in ferroelastic

ones. In addition to the thickness-dependent evolution of domain size, the domain pattern also changes with the film thickness, in particular, the gradual transformation from a ferroelastic domain structure to a ferroelectric one, and leads to the divergence of domain size scaling from conventional $1/2$ power law relationship.

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