Nonlinear Dynamics of Domain-Wall Propagation in Epitaxial Ferroelectric Thin Films

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We investigated the ferroelectric domain-wall propagation in epitaxial $Pb(Zr, Ti)O_3$ thin film over a wide temperature range (3–300 K). We measured the domain-wall velocity under various electric fields and found that the velocity data is strongly nonlinear with electric fields, especially at low temperature. We found that, as one of surface growth issues, our domain-wall velocity data from ferroelectric epitaxial film could be classified into the creep, depinning, and flow regimes due to competition between disorder and elasticity. The measured values of velocity and dynamical exponents indicate that the ferroelectric domain walls in the epitaxial films are fractal and pinned by a disorder-induced local field.

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The physics of surface growth in disordered media with quenched defects is of crucial importance to understand numerous intriguing natural phenomena [1], including contact lines in wetting, surface of epitaxially grown films, and magnetic domain walls. In such media, elastic forces tend to keep surfaces flat, while defects locally promote the wandering, as schematically displayed by Fig. 1(a). The competition between elastic and pinning forces leads to a complicated energy landscape with many local minima, which affects the surface growth dynamics under an external force. Recently, there have been extensive reports to adapt the fractal concepts to surface growth dynamics [2].

Ferroelectric (FE) domains have been studied for past decades because of scientific importance in microscopic aspects such as multidomain formation, stability, and pattern at equilibrium as well as technological applicability in multifunctional devices such as FE random access memories, actuators, and sensors [3,4]. Quite recently, lots of piezoresponse force microscope (PFM) studies have provided us microscopic aspects of FE domains, including inhomogeneous nucleation process [5] and the fractal nature of their rough surfaces [6]. Note that most works on FE domains have been focused on their static properties. In spite of its scientific and technological importance, we have limited understandings on how the FE domain-wall propagates in terms of time.

We suggest to prospect FE domain-wall from the view of nonlinear responses, which follow the predictions of the statistical physics on surface growth. Then, the FE domain-wall velocity v should have a nonlinear behavior, shown in Fig. 1(b), under E. At zero temperature T, the domain-wall remains strongly pinned by local disorders until E reaches a threshold value E_{C0}. When $E \geqq E_{C0}$, it experiences a pinning-depinning transition and starts to move with a nonzero velocity v, as represented with the red dashed line. Under this depinning regime,

$$v \sim (E - E_{C0})^{\theta}, \quad (1)$$

with a velocity exponent θ. Under the flow regime, when $E \gg E_{C0}$, $v \sim E$. On the other hand, for finite T, the pinning-depinning transition becomes relatively smooth, as represented with the green solid line. Under the low-E (i.e., $\ll E_{C0}$) creep regime, the domain-wall motion becomes very slow and can be described by propagation between pinning sites due to thermal activation. Then,

$$v \sim \exp[-(U/k_B T)(E_{C0}/E)^{\mu}], \quad (2)$$

where U is an energy barrier and μ is a dynamical exponent. Critical exponents of domain dynamics, including μ and θ, can identify the universality class and provide information on the pinning forces and the fractal nature of the rough FE domain walls. Although measurements of these critical exponent values for the FE systems are particularly important, there are little experimental works on these values.

In this Letter, we report our studies on the T- and E-dependent nonlinear responses of FE domain-wall dy-

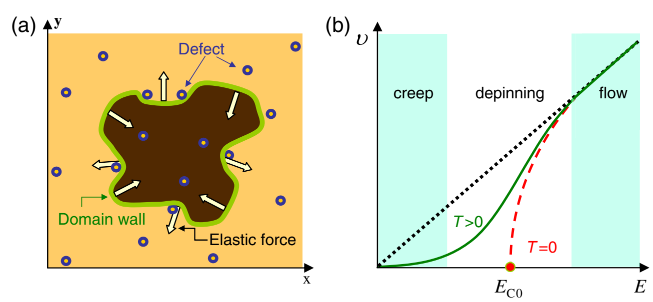

FIG. 1 (color online). (a) A schematic diagram of domain-wall propagation in disordered medium. Elastic forces come from the curvature of domain wall, and defects work as strong pinning sites. (b) Theoretical prediction on the domain-wall velocity v vs electric field E in system governed by competition between disorder and elasticity effects. E_{C0} represents a threshold E.

namics in epitaxial Pb(Zr, Ti)O_3 (PZT) thin film. To widen the accessible region of T and E, we used switching current measurements, combined with direct v data determined by PFM images. We found that v follows the nonlinear dynamic response, as described in Fig. 1(b). We also could obtain values of the two critical exponents, μ and θ, from the data in the creep and the depinning regimes, respectively. This work provides us new insights on how domain walls propagate inside epitaxial FE thin films.

We fabricated 100 nm-thick epitaxial PZT thin film on $SrRuO_3/SrTiO_3$ substrate using pulsed laser deposition [7]. X-ray diffraction studies confirmed that a high-quality, (001)-oriented PZT film was grown epitaxially. To fabricate PZT capacitors, we patterned the sputtered Pt top electrodes with a typical area of $7.5 \times 10^3\ \mu\text{m}^2$. Our epitaxial film revealed a high dielectric stability, suitable to our measurements at high electric fields.

Figure 2(a) shows T-dependent polarization–electric field (P-E) hysteresis curves for a PZT capacitor, measured between 3 and 300 K. The saturation and remnant P values were nearly constant over a wide T range. The systematic T variation of the P-E hysteresis curves comes mostly from T-dependent change in coercive field E_C. At 3 K, $E_C \approx 1$ MV/cm. As T increased, E_C decreased significantly.

One of the difficulties in performing the dynamic studies on FE domains is to measure reliable values of v under uniform E. Recently, we developed a modified PFM technique for a FE thin film with a top metal electrode [5,8]. By combining PFM with switching current measurements, we were able to track wall motions of domains. After applying one positive poling pulse (10 V, 50 μs) to pole P, we switched P with a series of negative pulses. We then measured the PFM images after all the negative pulses. We assumed that the PFM image obtained after the negative pulses would be nearly the same as that obtained after a single pulse, with the width being equal to the sum of all the negative pulses [8]. Typically, an image acquisition process using the PFM setup takes a few minutes, which is relatively long in relation to the time scale corresponding to the width of the E pulse used in these experiments. To check its validity, we calculated the amount of normalized switched polarization Δp from PFM images with an area of $5 \times 5\ \mu\text{m}^2$. These are shown as the open symbols in Fig. 2(b). We also determined Δp independently from switching current measurements with reading pulses of 50 μs, which are shown as the solid symbols. The agreement between these data indicates that we could reliably use the modified PFM techniques to study domain-wall motion in our FE film.

Figure 2(c) shows typical PFM images of time-dependent growth for a particular isolated domain, which were obtained at room T with $E = 0.3$ MV/cm. As time t increased, the domain size increased. Eventually, it began to merge with other domains. From the area of the isolated domain, we determined its mean radius. By dividing the change in mean radius with the change in t, we obtained the v value. We repeated these procedures for more than 20

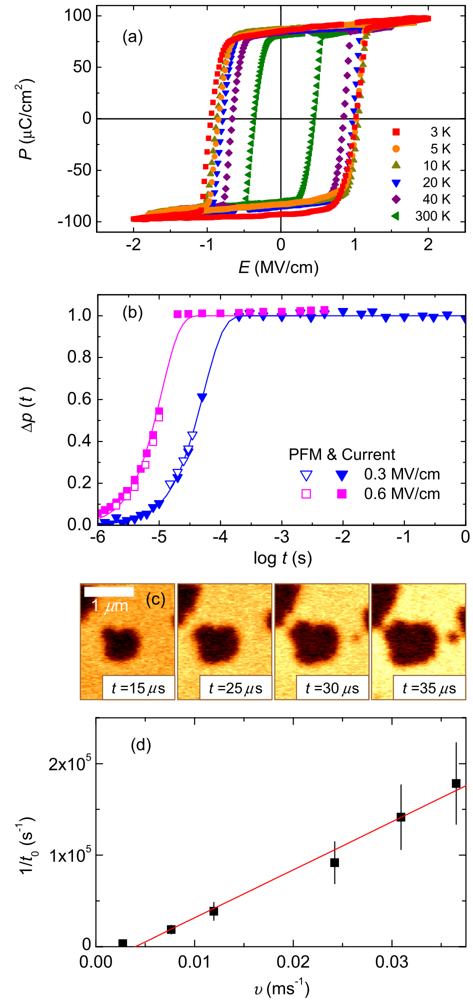

FIG. 2 (color online). (a) P-E hysteresis loops at different T. (b) Time-dependent normalized switched polarization, $\Delta p(t)$, behaviors (solid symbols) from switching current and (open symbols) from PFM measurements. The solid lines show the fitting results using the KAI model. (c) Scanned images of t-dependent domain growth at $E = 0.3$ MV/cm. (d) Plot of $1/t_0$ vs v. This relationship is linear, suggesting that $1/t_0$ from switching current studies can be used to parameterize v.

isolated domains and finally obtained an average value for the given T and E. Note that our modified PFM study can provide a way to measure v directly. However, obtaining sufficient data to plot all v vs E curves would have been too laborious. In addition, it is rather difficult to use in the low T region. Therefore, it is highly desirable to find another method to obtain v reliably in a wide range of T and E.

The switching current in a FE thin film should be originated from the P reversal, whose behavior is governed by domain-wall dynamics. The t-dependent changes in Δp for epitaxial FE thin films [9] have been explained using the Kolmogorov-Avrami-Ishibashi (KAI) model [10]. According to the classical statistical theory on nucleation and unrestricted domain growth,

$$\Delta p(t) = 1 - \exp[-(t/t_0)^n], \quad (3)$$

where n and t_0 are a geometric dimension and a characteristic switching time for the domain growth, respectively. In the two simplest cases, analytical relationships between t_0 and v can be easily obtained. When the nuclei of opposite polarity are generated at a constant rate under E, $t_0 \sim (1/v)^{(n-1)/n}$ [10]. Conversely, when all the nuclei are generated instantaneously, $t_0 \sim 1/v$ [10]. Our previous studies on epitaxial PZT films showed that nucleation rate is approximately proportional to $1/t$ [5], which is much closer to the latter case, so $1/t_0$ might be nearly proportional to v.

We investigated the relationship between v and $1/t_0$ using the switching current response. Our experimental $\Delta p(t)$ data were fitted using the KAI model, i.e., Eq. (3), as displayed by the solid lines in Fig. 2(b). This prediction was closely correlated with the $\Delta p(t)$ data. We compared $1/t_0$ values with v values, which were directly measured by PFM. As shown in Fig. 2(d), $1/t_0$ is linearly proportional to v with a small offset, indicating that the switching current response could provide reliable values of v.

Figure 3 shows $\Delta p(t)$ data with varying values of E. The solid symbols and lines indicate the experimental data and the fitting results using Eq. (3), respectively. At 300 K, a sudden change in $\Delta p(t)$ with $E = 0.1$ MV/cm occurred between 0.1 and 1.0 s: namely, at $t_0 \sim 2 \times 10^{-1}$ s. As E increased, a sudden change in $\Delta p(t)$ occurred over a shorter time scale, i.e., a smaller value of t_0. Conversely, at 3 K, little changes were observed in $\Delta p(t)$ when $E \leq 0.8$ MV/cm. This implied that the FE domain was pinned by defects and could not move below a threshold value of E. However, $\Delta p(t)$ starts to change abruptly around $E = 1$ MV/cm. This corresponded to the pinning-depinning transition. When E increased, t_0 rapidly decreased. The $\Delta p(t)$ data for 3 and 300 K were similar at $E = 1.1$ MV/cm; $t_0 \sim 5.4 \times 10^{-6}$ and 3.3×10^{-6} s at 3 and

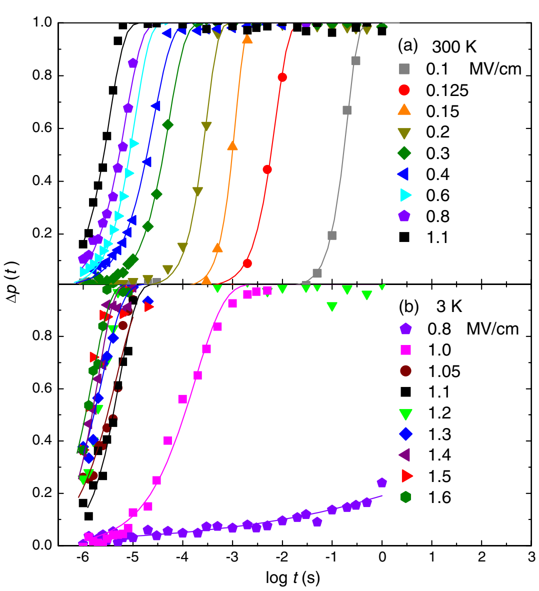

FIG. 3 (color online). $\Delta p(t)$ obtained via switching current measurements under various E values at (a) 300 and (b) 3 K. The solid lines show the fitting results using the KAI model.

300 K, respectively. These similar t_0 values demonstrated that the T dependence of v became insignificant under very high E region.

Figure 4(a) shows experimental $1/t_0$ vs E curves. Note that these $1/t_0$-E plots resemble those in Fig. 1(b). In the low-E region, $1/t_0$ was strongly dependent on T, consistent with predictions for the thermally activated creep regime. However, in the high E region, the values of $1/t_0$ started to merge, indicating a crossover to the flow regime. The pinning-depinning transition at 3 K occurred at approximately 1 MV/cm, which was close to the E_C value obtained from P-E hysteresis curve in Fig. 2(a).

From the experimental $1/t_0$ data, we obtained the value of the dynamical exponent μ for the creep regime. From Eq. (2), $\log(t_0)$ should be proportional to $(U/k_BT) \times (E_{C0}/E)^{\mu}$. The $\log(t_0)$ vs $1/E$ curves are shown in

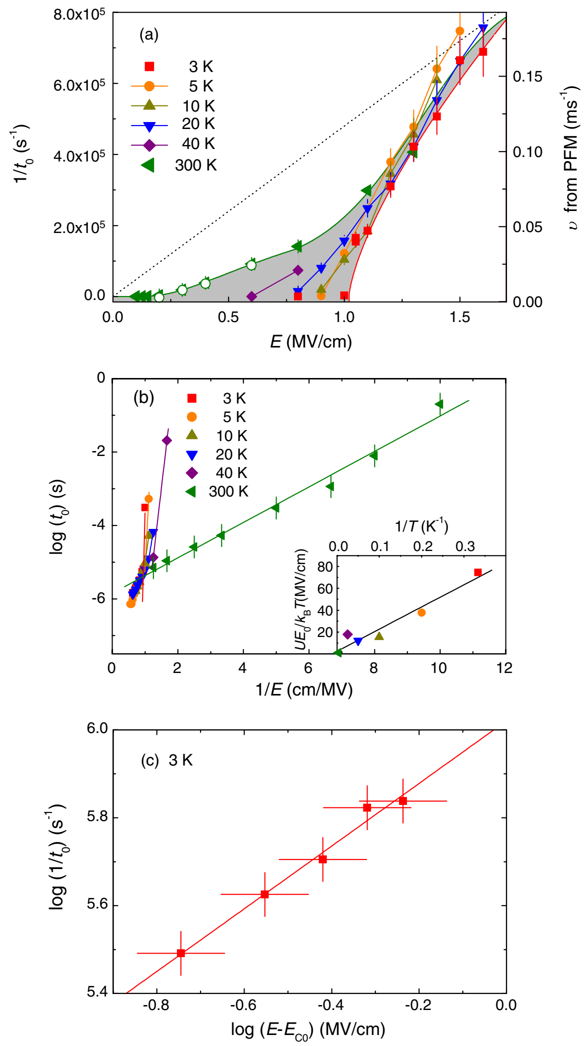

FIG. 4 (color online). (a) (Solid symbols) T dependent curves of $1/t_0$ vs E. The dotted and solid lines are guidelines for eye. (b) $\log(t_0)$ vs $1/E$ curves. The linear fitting indicates a dynamic exponent $\mu \sim 1$. The inset shows that UE_{C0}/k_B is nearly independent of T. (c) Plot of $\log(1/t_0)$ vs $\log(E - E_{C0})$ with the $E_{C0} \sim 1$ MV/cm. From the slope of the plot, we found a velocity exponent $\theta \sim 0.7$.

Fig. 4(b). The experimental data at a given T fall approximately into a linear line, indicating that μ is close to 1.0. Considering experimental errors, we found $\mu = 0.9 \pm 0.1$. This μ value agrees with previously reported values for epitaxial and polycrystalline PZT films [11,12]. The inset shows the value of $UE_{C0}/k_B T$ calculated from lines of best fit for several values of T. The straight line indicates that the value of UE_{C0}/k_B is nearly independent of T and is about 300 K MV/cm, close to previously reported value of around 400 K MV/cm from local domain switching data using PFM at room T [11].

The value of μ reflects the nature of the pinning potential in our PZT films. Under a pinning potential with a short range (the so-called random bond), one- and two-dimensional domain walls should have the μ values of 0.25 and 0.5, respectively [13]. However, under a pinning potential with a long range (the random field), $\mu = 1.0$ regardless of dimensionality [13]. Recently, two conflicting μ values were reported for epitaxial PZT thin films; 1.0 and 0.5–0.6 [11,14]. Our study confirms that $\mu = 0.9 \pm 0.1$ over wide range of T. This μ value suggests that the defects in our PZT thin film induce a long-ranged local field and pin FE domain walls [15].

From our experimental $1/t_0$ data, we also obtained the value of the velocity exponent θ near the pinning-depinning transition. By Eq. (1), $\log(1/t_0)$ should be proportional to $\theta \cdot \log(E - E_{C0})$. As shown in Fig. 4(c), this is approximately the case when $T = 3$ K. The slope of the line for best fit was $\theta \approx 0.71 \pm 0.05$. We could not find any earlier studies with which compare this θ value for FE thin films.

The θ value should reflect the dimensionality D of the surface for an elastic object in a disordered medium, as $\theta = (5 + D)/9$ [13]. Using this relationship, we found that $D \approx 1.4 \pm 0.4$ for our PZT thin film. Recently, several studies have reported noninteger dimensionality of local and static domain walls in FE thin films [6,16]. By measuring the local switching of PZT-$BiFeO_3$ sol-gel thin films with liquid PFM, Rodriguez *et al.* reported that $D \approx 1.5 \pm 0.1$ [6]. By measuring morphology and scaling of the domains in epitaxial $BiFeO_3$ thin films, Catalan *et al.* reported that $D \approx 1.5 \pm 0.1$ [16]. Our measured value of θ from the dynamic responses is consistent with these D values from the static measurements. In addition, our findings indicate that the newly observed fractal dimensionality of FE domains should be originated from local quenched defects.

We want to point out future studies and possible implications of our works: (i) in order to fully understand the domain-wall dynamics in epitaxial FE thin films, it is essential to measure other critical exponents, related to the divergence of the correlation length, the local variance of the domain-wall position, and so on [2]; (ii) domain dynamic responses in systems with different structural conditions such as polycrystalline FE thin films and ultrathin films should be investigated and compared with those in epitaxial films; (iii) the complete understanding on FE domain dynamics is necessary for numerous practical applications, including optimizing operation speed of miniaturized FE devices.

In summary, we investigated domain-wall motions of epitaxial PZT film over a wide range of temperature and applied electric field. We found that the motions were likely to be governed by the nonlinear dynamics of surface growth in a disordered medium with quenched defects. We determined two critical exponents for domain-wall propagation dynamics, which indicate the random field nature of the defects and fractal nature of domain walls.

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