Surface effects on switching in ferroelectric films

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Surface effects on switching behaviours of ferroelectric (FE) films are elucidated by using the Landau Devonshire (LD) free energy expression. The well-known parameter, δ, the extrapolation length, introduced in the LD free energy expression strongly influences the switching behaviours in FE films. Polarization at the surface can either be suppressed when δ is positive or be enhanced when δ is negative. We have discovered that the delay in switching at the centre relative to switching near the surfaces is more remarkable in the case of $\delta = 0$ than the non-zero δ cases, as the film thickness increases. It is also found that the reversals of dipole moments at the centre and near the surface, by the applied field, take place almost together in FE thin films of non-zero δ. The results we obtained have shown that the surface extrapolation length δ also influences the critical thickness of FE films.

I. INTRODUCTION

Polarization reversal in ferroelectric (FE) materials has been an important area of study in ferroelectricity for a long time since some significant measurements on switching behaviours of FE crystals have been made by Merz and other workers [1,2] about half a century ago. The interest in this research area has further extended to FE thin films; and the interest has not waned even up to these days because of the advancement in thin film fabrication technology, where higher quality and more reliable FE thin films can be fabricated; thus making applications of FE thin films in microelectronic devices and memories [3-5] more reliable. More current theoretical and experimental elucidations in polarization reversal in FE thin films are focused more on phenomena related to effects of size and surface in thin films on switching time and coercive field [6-10]. From the literature, on theoretical studies, several models based on a Landau-typed phase transition have given well explanations on switching behaviours of mesoscopic ferroelectric structures [7,9]; and some of the predictions concerning size and surface effects on switching behaviours by Landau-typed models agree well with experimental observations. However, there still lacks detailed understanding of surface effects on FE films under the applied electric field, which is a very important portion of knowledge toward the overall understanding on the behaviours of polarization reversal in the films as a whole. In this paper, we adopt the Tilley-Zeks model of Landau Devonshire (LD) free energy expansion to study the effect of surface conditions under the applied electric field on a one dimensional FE film. With this framework, we present the results of switching behaviours of FE film with zero and non-zero values of δ as surface conditions.

II. THEORETICAL MODEL

A symmetric second-order FE thin film of thickness L in which the polarization and related dielectric quantities vary as a function of z is considered. The film thickness considered is much smaller than the lateral dimension, thus depolarization effects can be neglected. In our case, the film thickness extends from one surface with $z = -L/2$ to another with $z = +L/2$; and the effects due to surfaces are governed by an extrapolation length δ [11]; where δ can be positive or negative. For positive value of δ, polarization is suppressed at the film surfaces and the reversed is true for negative δ. Usual LD model used in elucidation of dynamic properties of FE films considers the effect of applied electric field E to the film without the surface potential term [10,12]. However, it has been proposed by Ishibashi *et al.* [13] that the surface potential term due to the effect of electric field has been included in the free energy expression. Since the surface thickness in the continuum model is zero, we have the opinion that the surface potential due to electric field may not be necessary to be considered in the free energy as many researchers have. With these conditions stated, the LD free energy per unit area of the film can be defined:

$$\frac{F}{S} = \int_{-L/2}^{L/2} f(P, P')dz + \frac{C}{2\delta\varepsilon_0}\left(P_-^2 + P_+^2\right) \qquad (1)$$

where

$$f(P,P') = \frac{1}{2\varepsilon_0}AP^2 + \frac{1}{4\varepsilon_0^2}BP^4 - EP + \frac{1}{2\varepsilon_0}CP'^2,$$

$P' = \dfrac{dP}{dz}$ and $P_{+,-} = P\left(\pm\dfrac{L}{2}\right)$. The parameters B and C are the usual second-order Landau coefficients, while $A = \alpha(T - T_0)$ with α is the inverse Curie constant and

T_0 is the bulk critical temperature. Minimization of Eqn. (1) by variational method shows that the polarization satisfies the Euler Lagrange (EL) equation

$$\frac{A}{\varepsilon_0}P+\frac{B}{\varepsilon_0^2}P^3-E-\frac{C}{\varepsilon_0}\frac{d^2P}{dz^2}=0 \tag{2}$$

with the following boundary conditions:

$$\frac{dP}{dz}=\pm\frac{P}{\delta} \quad \text{at} \quad z=\pm L/2\,. \tag{3}$$

For the purpose of numerical calculations, Eqn. (2) and Eqn. (3) are scaled to dimensionless units in the normal way [16], and we have

$$(t-1)p+p^3-\frac{2}{3\sqrt{3}}e-\frac{d^2p}{d\zeta^2}=0 \tag{4}$$

$$\frac{dp}{d\zeta}=\pm\frac{p}{d} \quad \text{at } \zeta=\pm l/2 \tag{5}$$

where $p=P/(\varepsilon_0\alpha T_0/B)^{1/2}$; $t=T/T_0$; $e=E/\left(\alpha^3T_0^3/\varepsilon_0 B\right)^{1/2}$; $\zeta=z/\xi_0$, $\xi_0^2=C/\alpha T_0$; $l=L/\xi_0$ and $d=\delta/\xi_0$. The length scales *L*, δ and λ are usually scaled according to ξ_0, the characteristic length of the material [10,11] and it can be related to the thickness of domain wall.

III. NUMERICAL CALCULATIONS

To discuss the dynamics of polarization reversal in the film, the Landau-Khalatnikov (LK) equation of motion is applied. As a consequence of omitting the kinetic term for low frequency studies, the dimensionless LK equation, following the scaling mentioned above, can be written as follow:

$$\frac{dp}{d\tau}=-(t-1)p-p^3+\left(\frac{d^2}{d\zeta^2}\right)+\frac{2}{3\sqrt{3}}e \tag{6}$$

where $\frac{dp}{d\tau}$ is the dimensionless switching current and τ ($\tau=t'/\frac{\alpha T_0}{\varepsilon_0 C}$; t' is the time in real unit) is the reduced time. Polarizations at the film surfaces are influenced by the extrapolation length δ during switching. Numerical process in simulation of switching profiles in various cases of surface conditions requires a static polarization profile as initial value for the LK equation and the exact solution from equations Eqn. (2) and Eqn. (3) has been obtained by Ong *et al*. [11]. When an electric field *e* ($e>e_{cf}$) is applied, LK equation in Eqn. (6) is used to simulate the profiles of polarization during switching.

The applied electric field used in the simulation takes the form of step function in reduced units as

$$e=e_0\theta(\tau)\,, \tag{7}$$

where e_0 is the amplitude, for $e_0=1$ gives the equivalent of the zero-temperature bulk coercive field e_c and $\theta(\tau)$ is the usual step function.

IV. RESULTS AND DISCUSSION

Various stages of switching profiles for zero and non-zero δ FE films are shown in Fig. 1 and Fig. 2 respectively for temperature *t* = 0.6. The starting equilibrium polarization profiles in both cases (Fig. 1 and Fig. 2) are set at negative at time $\tau=0$; and the profiles are switched by a positive applied electric field *e* described in Eqn. (7) until it is completely saturated at $\tau=\tau_s$. τ_s is the switching time, and it is defined as the time taken for the polarization current to decline to 10% of its maximum value [7,14].

The general trend in polarization reversal of a film, irrespective of value of δ, shows clearly that switching time τ_s is longer as the film gets thicker (Fig. 1 and Fig. 2). Figs. 1(a) and (b) show the switching profiles of a thinner film (*l* = 3.3) of $\delta=0$ and non-zero δ where all parts of the profiles are shown to switch almost together. There is not much difference in the switching speed between regions near the surface and the interior of the film. In a thicker film (*l* = 7.0), the reversal of polarization begins near the surfaces first, and then goes on to the centre, as shown in Figs. 2(a) and (b). This indicates that the domain wall is formed near the surfaces, followed by domain wall movement towards the centre. Hence, when the film becomes thicker, the delay in switching at the centre of the film is more distinct compared with the delay nearer the film surfaces. In term of domain wall movement in the film, it obviously takes a longer time for a domain wall to move from the surface to the entre for a thick film than a thin film. It can be seen that the delay in switching at the centre in a zero-δ film is more remarkable when the film becomes thicker (Fig. 3).

Fig. 4 shows the average polarization of a FE film versus thickness for various values of δ. It is clear that the extrapolation length δ has great effects on the critical thickness; and the decrease in critical thickness l_c with increasing value of δ that has been predicted in ref. 11. In addition, the coercive field e_{cf} of a free boundary film increases with the value of δ. As for a FE film with larger δ, pinning at the surfaces is weak, resulting in a larger surface polarization. Consequently, a larger field is required to reverse the polarization.

V. CONCLUSIONS

The effect of surface conditions, due to δ, on switching behaviours of FE films is studied by using the continuum LD model. It has been shown that switching at the surface and at the centre is almost the same in thin FE films irrespective of δ. For thicker films, surface switching takes place relatively faster than the interior of the films. However, the delay in switching at the centre is more remarkable in the zero-δ film as the film thickness increases.

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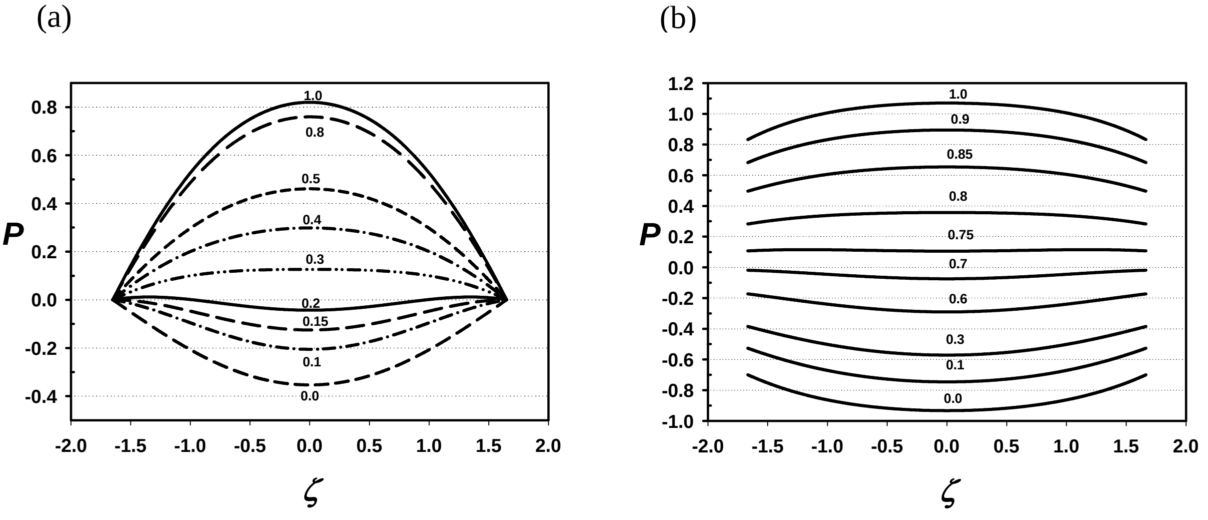

FIG. 1. Polarization profiles during switching, at various time in term of fraction of the switching time τ_s, at temperature t = 0.0, applied field e = 0.83, thickness l = 3.3 for (a) δ = 0; (b) δ = 2.0. The number at each curve represents time taken to reach the stage in term of fraction of τ_s.

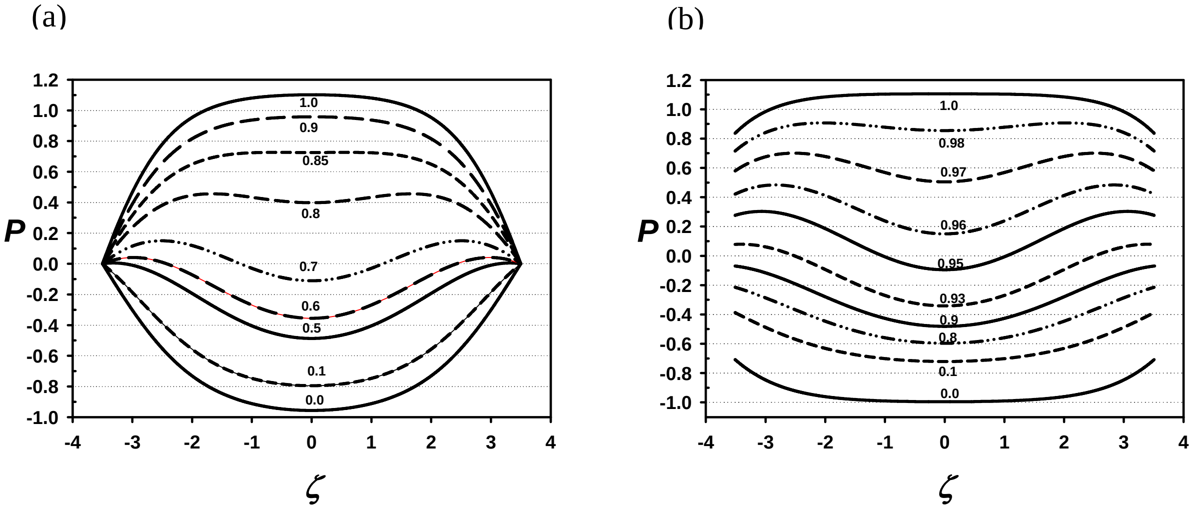

FIG. 2. Polarization profiles during switching, at various time in term of fraction of the switching time τ_s, at temperature t = 0.0, applied field e = 0.83, thickness l = 7.0 for (a) δ = 0; (b) δ = 2.0. The number at each curve represents time taken to reach the stage in term of fraction of τ_s.

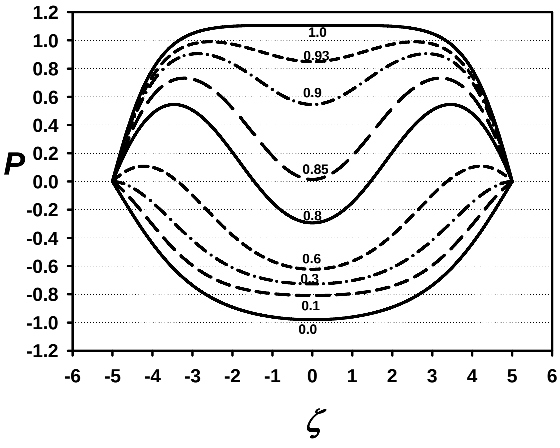

FIG. 3. Polarization profiles during switching, at various time in term of fraction of the switching time τ_s, at temperature t = 0.0, applied field $\boldsymbol{e}$ = 0.83, thickness l =10.0, δ = 0.

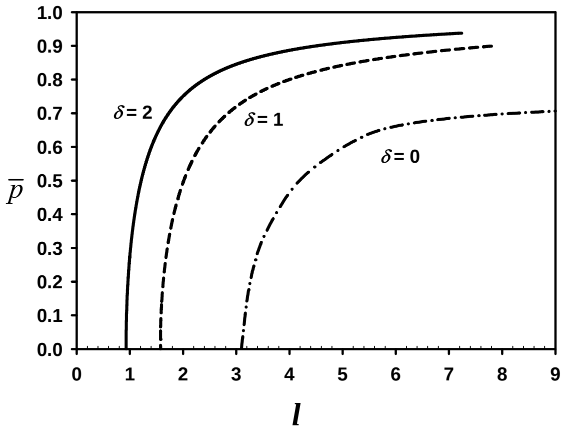

FIG. 4. Average polarization versus thickness, l for various values of δ.