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Dynamic process of domain switching in ferroelectric films

Biao Wang[a)] and R. Xia
Electro-Optics Technology Center, Harbin Institute of Technology, Harbin, 150001, China

H. Fan
School of MPE, Nanyang Technological University, Singapore 639798

C. H. Woo
Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hong Kong, China

In this article, a general variational formulation is established for the dynamics of domain switching in ferroelectric thin films. By incorporating the kinetic energy in the Landau free energy expansion, we obtain a second-order time-dependent Ginzburg–Landau evolution equation, which can be used to analyze the frequency-dependent dielectric and ferroelectric properties of the materials. A numerical model is developed to calculate the time-dependent polarization and electric-field distribution during the domain switching process. As an example of practical importance, a ferroelectric film with 180° domain boundaries perpendicular to the film surface is studied. Domain switching is found to proceed in two stages, the initial nucleation and growth of the ferroelectric domain, followed by the expansion of the domain through the sidewise motion of the 180° domain walls, until the spontaneous polarization in the entire electrified area is reversed. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1599958]

I. INTRODUCTION

Besides obvious scientific interest, ferroelectric crystals have been a subject of intense investigation for several decades because of their many possible interesting applications to electronic and electro-optical devices, which are derived from the fact that the polarization and distortion of these crystals can be altered via the action of an applied electric or mechanical field, and vice versa.

Above the Curie temperature, ferroelectric crystals are nonpolar, but below, they are spontaneously polarizable by the distortion of the lattice. As a result, a ferroelectric crystal below the Curie temperature generally has a structure consisting of many uniformly oriented spontaneously polarized regions, i.e., ferroelectric domains. In this regard, a wall between domains with opposite oriented polarizations is a 180° wall, and a 90° wall, if the polarizations it separates are mutually perpendicular. Ferroelectric domains can be switched and domain patterns can be manipulated by means of an applied electric field. Since domain switching with its large change of polarization and lattice distortion provides a potent mechanism for many interesting properties, a clear understanding of the domain switching process in ferroelectrics is important.

To investigate the polarization distribution and domain structures, a conventionally effective method is to establish a phenomenological model based on the Landau free energy expansion.[1–3] One can then establish both the time-dependent and time-independent Ginzburg–Landau equations with respect to the polarization. Unfortunately, the re-

sulting classical Ginzburg–Landau equations are not applicable in the case of frequency-dependent properties of ferroelectric materials. Thus, it is well known, both from the lattice dynamics analysis[4] and from experiments,[5,6] that the dielectric response of ferroelectric materials to a high-frequency applied electric field has a resonance peak. The classical time-dependent Ginzburg–Landau equation, being a first-order temporal differential equation, cannot produce this kind of dynamic response.

The dynamics of the dielectric response of ferroelectrics has become a subject of increasing attention because of applications in high-frequency electronic devices, such as static memories, sensors, microstrips lines, etc. By postulating the presence of inertia during polarization switching in the Landau free energy expansion, we introduce, in this article, a kinetic energy term that is proportional to the square of the polarization switching speed. In this way, a second-order temporal evolution equation is obtained. By considering the harmonic part of the equation, the dynamic dielectric response of the ferroelectric material under the action of an oscillating electric field can be calculated.

Periodic or quasi-periodic structures,[7] consisting of metallic or semiconducting layers, often possess special physical properties, such as localization of electronic states[8] and high efficiency of energy conversion, upon which their functionality in various electronic devices depends. In general, there are two types of periodic domain structures with antiparallel polarization, namely, those formed with the "head-to-tail" orientation [Fig. 1(a)], and others with "head-to-head" orientation [Fig. 1(b)]. In addition, there is also the quasi-periodic domain structure (i.e., quasi-periodic superlattices),[9] in which the building blocks are 180° domains, but differ somewhat in size [Fig. 1(c)]. In the follow-
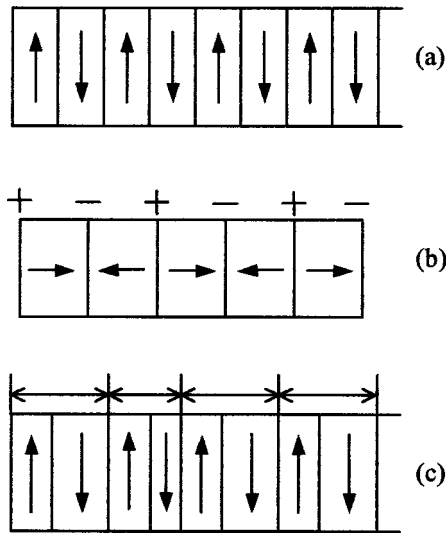
FIG. 1. Different domain structures: (a) head-to-tail, (b) head-to-head, and (c) a quasi-periodic.

ing sections, numerical calculations are carried out on a LiNbO$_3$ (Fig. 2) ferroelectric film with periodic domain structures.

II. ENERGY CONCEPT OF DOMAIN STRUCTURE AND FORMULATION

Consider a ferroelectric thin film with the dimension $x \times y \times z = L_x \times L_y \times h$, with h being the film thickness. The origin of the coordinate system is at the center of the cell. The nonzero component P of the spontaneous polarization of the ferroelectric thin film is assumed to be orthogonal to the surface of the film. In general, including the effect of depolarization field $\vec{E}_d = (E_x, E_y, E_d)$, the applied electric field $\vec{E}_{\text{ext}} = (0,0,E_{\text{ext}})$, and the epitaxial stress $\sigma_{xx} = \sigma_{yy} = \sigma_r$ in the ferroelectric thin film, one can write the free energy of the cell as
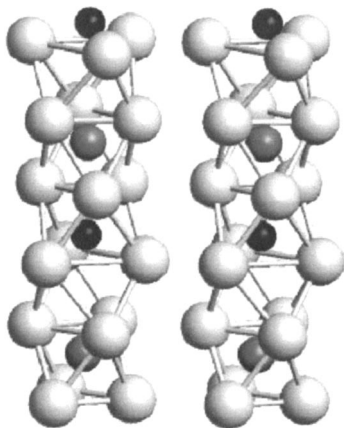

FIG. 2. Stereoscopic view of the ideal crystal stacking sequence of lithium niobate along the crystallographic c axis (light gray: Oxygen, dark gray: Niobium, and black: Lithium).
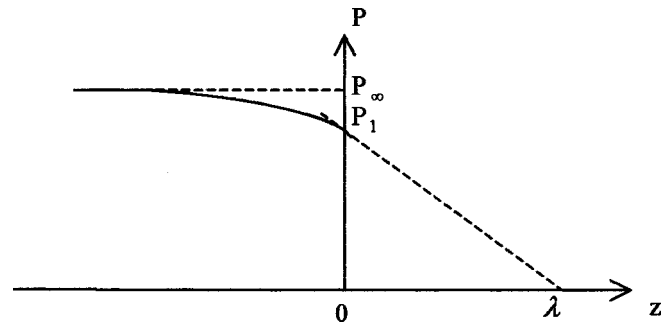

FIG. 3. Variation of the local polarization in the vicinity of a film free surface situated $z=0$. The bulk polarization is denoted by P_∞ while the P_1 denotes the polarization at the surface.

$$
F = \int \int \int_V \left\{ \frac{A}{2} P^2 + \frac{B}{4} P^4 + \frac{C}{6} P^6 + \frac{D_{11}}{2} \left(\frac{\partial P}{\partial z} \right)^2 \right.
$$
$$
+ \frac{D_{44}}{2} \left[\left(\frac{\partial P}{\partial x} \right)^2 + \left(\frac{\partial P}{\partial y} \right)^2 \right] - \frac{1}{2} E_d P - E_{\text{ext}} P
$$
$$
\left. - 2\sigma_r \varepsilon_0 \right\} dv + \int_{S_1} \int \frac{D_{11} \delta_3^{-1}}{2} P^2 dx dy
$$
$$
+ \int_{S_2} \int \frac{D_{44} \delta_1^{-1}}{2} P^2 dy dz + \int_{S_3} \int \frac{D_{44} \delta_1^{-1}}{2} P^2 dx dz,
$$

$$(1)$$

where A, B, C, D_{11}, and D_{44} are expansion coefficients of the corresponding bulk material, S_1, S_2, *and* S_3 represent the six surface planes, of the cell. δ_i is the extrapolation length, and measures the strength of the surface effect (Fig. 3),[10,11] and ε_0 is the stress-free strain due to the phase transition, which is given by[1]

$$
\varepsilon_0 = \varepsilon_{xx}^T = \varepsilon_{yy}^T = QP^2, \tag{2}
$$

where Q is the electrostrictive coefficient.

Equation (1) is a classical form of the Landau free energy expansion by adding the contributions of the film surface, depolarization field, and epitaxial stresses. From the atomistic point of view, the polarization process is related to the noncentrosymmetric arrangement of ions.[4] If domain switching can be assumed to be a dynamic process, there should be a kinetic energy term, expressed as

$$
T = \frac{1}{2} \rho \left(\frac{\partial P}{\partial t} \right)^2, \tag{3}
$$

where ρ is a material parameter having the meaning of the effective mass density.

The evolution of the system can be determined based on Hamilton's principle. Let us define the Lagrange function as

$$
L = \int \int \int_V T dv - F. \tag{4}
$$

The dynamic behavior of the system between time t_1 and t_2 then depends on the "*action*" integral I,

$$I = \int_{t_1}^{t_2} L\,dt = \int_{t_1}^{t_2} (T-F)\,dt. \tag{5}$$

Similar to the time-dependent Ginzburg–Landau equation, one can then establish the polarization evolution equation based on the action integral as follows,

$$K\frac{\partial P}{\partial t} = \frac{\delta I}{\delta P}$$

$$= -\rho\frac{\partial^2 P}{\partial t^2} + \left\{ -AP + 4Q\sigma_r P + E_d + E_{\text{ext}} - BP^3 \right.$$

$$\left. -CP^5 + D_{11}\frac{\partial^2 P}{\partial z^2} + D_{44}\left(\frac{\partial^2 P}{\partial x^2} + \frac{\partial^2 P}{\partial y^2} \right) \right\}, \tag{6}$$

where K is the kinetic coefficient related to the domain-wall mobility. In deriving Eq. (6), the depolarization field is considered a linear function of the polarization. The divergence theorem for the electrostatic depolarization potential Φ can be written in the form,

$$\frac{\partial^2\Phi}{\partial x^2} + \frac{\partial^2\Phi}{\partial y^2} + \frac{\partial^2\Phi}{\partial z^2} = \frac{1}{\alpha}\frac{\partial P}{\partial z}, \tag{7}$$

where α is the dielectric constant of the film without the effect of spontaneous polarization, and

$$E_d = -\frac{\partial\Phi}{\partial z}. \tag{8}$$

If we neglect the nonlinear terms in Eq. (6), the applied electric field varies as $E_0\exp(-i\omega t)$, and the polarization $P = P_0\exp(-i\omega t)$ keeps spatial uniformity, and $E_d = -P/\alpha$, the dielectric susceptibility is given by

$$\varepsilon(\omega) = \frac{P}{E} = \frac{1}{1/\alpha + A - 4Q\sigma_r - \rho\omega^2 - iK\omega}. \tag{9}$$

The dielectric susceptibility in Eq. (9) has a real and an imaginary component, and a resonance is expected at the appropriate frequencies, in qualitative agreement with the experimental observations.[5,6] On the other hand, if we neglect the second-order time derivative in Eq. (6), Eq. (9) becomes the classical time-dependent Ginzburg–Landau equation, and this behavior of the dielectric susceptibility would be absent. Since the parameter A is negative below the Curie temperature, and the epitaxial stresses can be assumed to be zero, the effect of the depolarization is crucial for the behavior of the dielectric susceptibility.

III. NUMERICAL SIMULATION RESULTS AND DISCUSSION

Analytic solutions of the polarization P, the electric potential ϕ_d and ϕ_{ext} from Eqs. (7)–(9) are difficult to obtain. Nevertheless, the evolution of the polarization and electric fields and, thus, the domain structures, can be obtained numerically.

A numerical calculation is performed in three-dimensional space with two spatial coordinates and one temporal coordinate. In this work, we use a 200×400 lattice with periodic boundary conditions along the two Cartesian
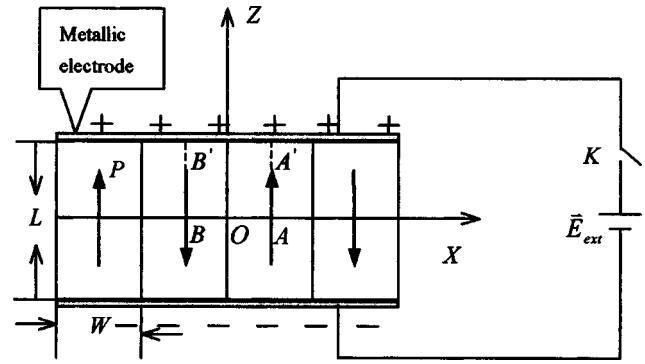

FIG. 4. Sketch of a ferroelectric single crystal film between two metallic electrodes in a dc electric field.

axes (Fig. 4). We note that the use of periodic boundary conditions imposes artificial periodicity of ferroelectric domains. This is particularly true when the domain size is comparable to the computational cell size.

In this calculation, the "damping term" is neglected, and a set of dimensionless variables[12] is adopted. Since A, B, D, ρ, and P have units of $\text{C}^{-2}\,\text{N}\,\text{m}^2$, $\text{C}^{-4}\,\text{N}\,\text{m}^6$, $\text{C}^{-2}\,\text{N}\,\text{m}^4$, $\text{C}^{-2}\,\text{N}\,\text{m}^2\,\text{s}^2$, and C/m^2, respectively, it is clear that $\sqrt{D/|A|}$ and $\sqrt{\rho/|A|}$ have dimensions of length and time, respectively. Therefore, a natural choice of dimensionless reduced variables is

$$\tilde{r} = \sqrt{|A|/Dr},$$

$$\tilde{t} = \sqrt{A/\rho t}, \tag{10}$$

$$\tilde{P} = P/|P_0|,$$

where P_0 is the spontaneous polarization at a given temperature. In the reduced variables, the effective coefficients are related to the original ones as follows

$$\tilde{A} = A/|A|,$$

$$\tilde{B} = BP_0^2/|A|, \tag{11}$$

where $\tilde{A} = -1$ (A is negative to make the ferroelectric phase stable). In the reduced variables, the lattice spacing in real space is chosen to be $\Delta\tilde{x} = 0.125$, $\Delta\tilde{z} = 0.25$, and the time step is $\Delta\tilde{t} = 0.006$.

To demonstrate the dynamics of the switching processes related to the reversal of permanent electric dipoles in a ferroelectric single crystal film, a sequence of images is shown, and the evolution of 180° periodic domain structure is represented as a function of time under the application of an external electric field whose magnitude is fixed along the negative z direction. In Fig. 5, the "color levels" represent different magnitudes of the polarization. A completely black region indicates that the polarization is equal to -1 and its direction is along the negative z direction; a light black region means that the polarization is equal to 1 and the polarization is along the positive z direction; the ferroelectric domain-wall region is revealed as a bright stripe, like the morphology of the cleavage. As shown in Fig. 5(a), at the initial moment $\tilde{t} = 0$ when there is no external electric field, the positive domain and the negative domain have uniform
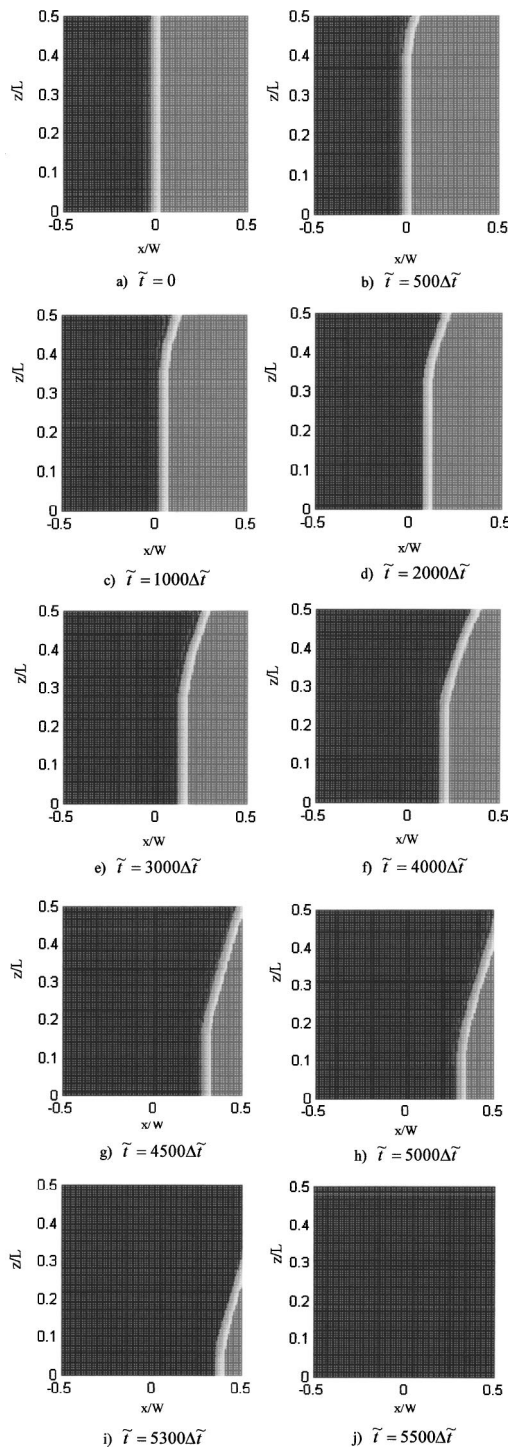
FIG. 5. Temporal evolution of 180° periodic domain under the application of external electric field: $\tilde{\alpha}_1 = -1$, $\tilde{\beta} = 1.0$, $W = 25$, $L = 100$, $E_{\text{ext}} = 5.0 \times 10^{-2}$, and $\Delta\tilde{t} = 0.006$.
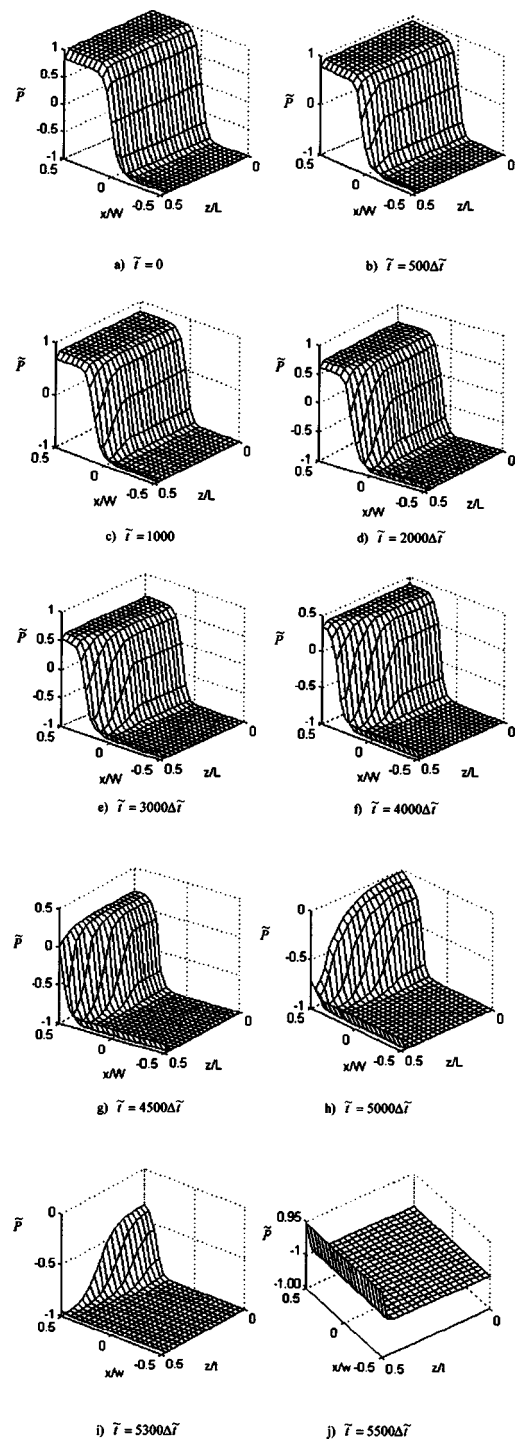

FIG. 6. The spatial distributions of the polarization in the rectangle $AA'B'B$ as a function time under the application of the external electric field: $\tilde{\alpha}_1 = -1$, $\tilde{\beta} = 1.0$, $W = 25$, $L = 100$, $E_{\text{ext}} = 5.0 \times 10^{-2}$, and $\Delta\tilde{t} = 0.006$.

fractions of volume and are separated by a domain wall parallel to the z axis. Figure 6 shows the variation of polarization along the z and x directions, and the spatial distribution of the depolarizing electric potential in the rectangle is represented in Fig. 7. The polarization near the film surface ($z = L/2$) is smaller than that in the interior of film. This is expected due to the surface effect and the strong depolarizing field arising from the negative gradient of depolarizing electric potential.

During the initial stage of domain switching, when the system is under the action of an external electric field [Figs. 5(b) and 5(c)], a domain with the shape of a triangle first forms near the electrode. Meanwhile, since the additional wall created during the nucleation event has to consume energy, nucleation along existing 180° domain walls becomes preferable. This result is in agreement with the theoretical assumption.[13] As the system evolves, the 180° domain wall moves along its normal; simultaneously, a negative domain
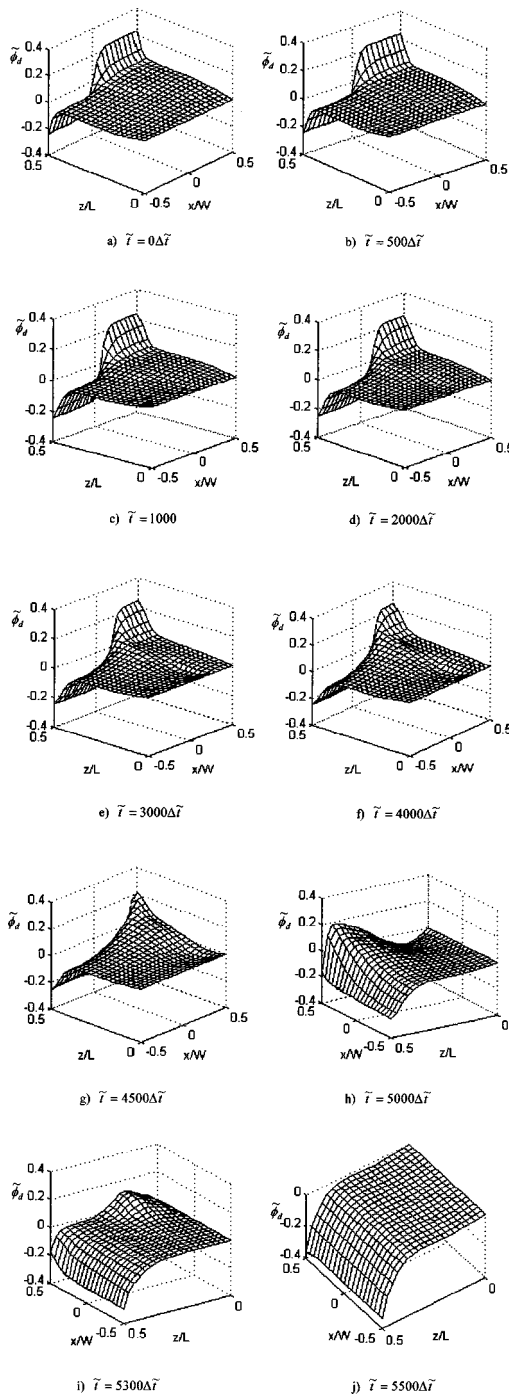
FIG. 7. The spatial distributions of the depolarizing electric potential in the rectangle $AA'B'B$ as a function time under the application of the external electric field: $\tilde{\alpha}_1 = -1$, $\tilde{\beta} = 1.0$, $W = 25$, $L = 100$, $E_{\text{ext}} = 5.0 \times 10^{-2}$, and $\Delta\tilde{t} = 0.006$.

expands itself into the interior of the film, and the fraction of positive domain decreases, as the fraction of negative domain increases. Eventually, as shown in Fig. 5(j), it takes about 5500 time steps until only the negative domain survives, i.e., the 180° domain wall disappears completely and the final equilibrium structure consists of a single domain. Figure 8 shows the dependence of the domain-wall velocity on the external electric field. The switching characteristics of the ferroelectric thin film may be estimated by using the
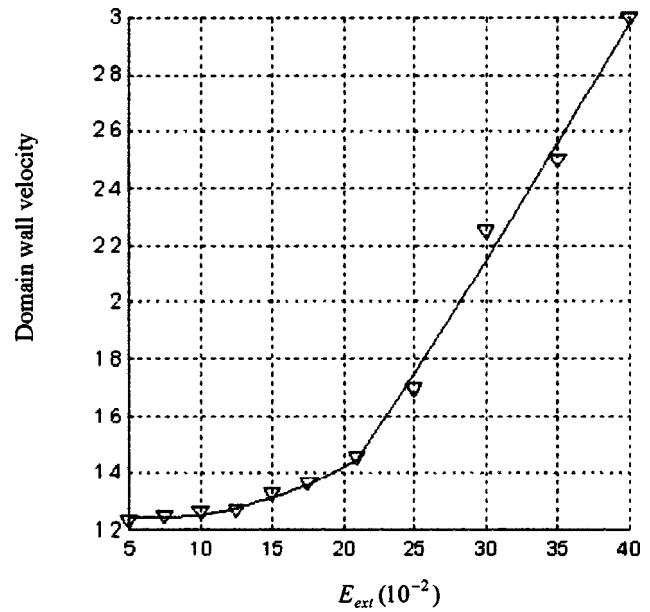

FIG. 8. Dependence of the domain-wall velocity on the external electric field.

curve in Fig. 8. As the external electric field is low, a non-linear dependence is shown in Fig. 8. As the external electric field increases, however, the dependence becomes linear.

IV. CONCLUSION

In the present article, a phenomenological, second-order evolution equation of polarization is established by incorporating an effective kinetic energy term with the Landau free energy expansion. Based on the equation, frequency-dependent dielectric properties of ferroelectric materials can be studied. As an example, the dynamic domain switching process in a ferroelectric thin film is considered. The results show that domain switching by an applied electric field with a direction opposite to the spontaneous polarization vector is a multistage process. The initial stage involves the nucleation near the electrode, and the growth along the existing domain wall, followed by the subsequent expansion through sidewise motion of the 180° domain wall, until a single domain is formed. By properly adjusting the duration of external electric field, the domain structure can be controlled.

ACKNOWLEDGMENTS

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J. Appl. Phys., Vol. 94, No. 5, 1 September 2003

Wang *et al.* 3389

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