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Calculations on switching characteristics of ferroelectric–paraelectric superlattices

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Abstract

We study the switching characteristics of a superlattice consisting of a periodic ferroelectric and paraelectric layer with interlayer coupling using the Landau–Khalatnikov equation. The inhomogeneity of polarization at the ferroelectric or paraelectric surface is described by the extrapolation length δ. The effects of various parameters such as the viscosity coefficient, layer thickness, interlayer coupling and temperature on the switching characteristics are investigated. Our study shows that the hysteresis loop and switching current versus applied field depend strongly on those parameters. Interlayer coupling increases the ferroelectricity of the superlattice, leading to a strong enhancement in polarization and coercive field.

Keywords: B. Interfaces; C. Ferroelectric properties; Polarization switching; Superlattices

1. Introduction

The efficiency in fabricating high quality layered structures of ferroelectric (FE) materials using advanced fabrication techniques and the increasing importance of FE nanostructures in FE memories, sensors, microelectro-mechanical (MEM) systems, optical devices, etc. have enhanced the motivation in experimental and theoretical studies of FE superlattices and multilayers [1]. However, the involvement of researchers in this area of research is still not flourishing. Experimental studies on oxide-material superlattices include the ferroelectric/ferroelectric (FE/FE) superlattices (for example, $BaTiO_3/PbTiO_3$ [2]), and ferroelectric/paraelectric (FE/PE) superlattices such as $BaTiO_3/SrTiO_3$ [3–5], $KNbO_3/KTaO_3$ [6–8], and $PbTiO_3/SrTiO_3$ [9–11] besides the FE multilayers and the solid solutions. It was discovered that physical properties of the artificially fabricated FE superlattices such as spontaneous polarization, the Curie temperature, and the dielectric susceptibility constant differ significantly from those of the simple bulks and thin films of the same FE materials [2–9]. From the experimental results reported, the dielectric constant of a designed superlattice is not a serial connection of the component layers [12] and the individual layer thickness of the whole structure can play an important role in the combined effect [5]. However, the underlying physics on how this effect comes about is still unclear, but it is simply related to the long-range dipole–dipole interaction within the coupled sublattices [1–11]. A better understanding of how the combination of those effects on the properties is very crucial in the synthesis of superlattices for particular applications [13].

The influence of intrinsic coupling, the interfacial coupling and strain, as well as the size effect on FE/FE and FE/PE superlattices are also theoretically elucidated by a few models including the Ising model in transverse field (IMTF) [14,15], thermodynamic model [16–18] and the shell model [19]. Through the theoretical studies, long-range interaction of dipoles, which causes the behaviours of FE/FE and FE/PE superlattices to be uniquely different from their constituent layer materials, is widely accepted [14–19]. In the thermodynamic model, the surface effect considered in the thin film is adopted, together with the interlayer/interfacial coupling that is used to study the role of long-range dipole interaction, which lead to the unique properties of FE/FE superlattices, in the coupled sublattices [13–19]. The experimental work on FE/FE and FE/PE superlattices are not many comparing with the studies in FE bulk and thin films; and it is even much lesser in theoretical studies on similar subject. From the limited theoretical studies, to the best of our knowledge, besides

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Sepliarsky et al. [19], very little work is done on the switching characteristics of the FE/FE and FE/PE superlattices.

In this study, we modified the well-established thermodynamic model developed by Tilley and Zeks for the thin film [20] to elucidate preliminarily the switching characteristics of a FE/PE superlattice of coupled sublattices formed from two second order materials. This model is an improved version to what we had proposed previously [16], but now the spatial variation of polarization is included in the free energy and naturally, the surface energy term will have to be included. The introduction of the surface energy into the free energy of the system brings in a parameter δ, called the extrapolation length. The long-range dipole–dipole interaction is governed by the coupling constant J, where $J>0$ brings along the antiferroelectric coupling on the coupled sublattices [16]. Polarization hysteresis and switching current obtained from the applied sinusoidal electric field by varying some parameters such as field strength, layer thickness, etc. are discussed.

2. Theory

We consider a periodic superlattice comprising a FE layer of thickness L_1 and a PE layer of thickness L_2, as shown in Fig. 1; and the periodic thickness of the superlattice is denoted as $L = L_1 + L_2$. The Landau–Ginzburg free energy per unit area F of the FE/PE superlattice is

$$F = F_1 + F_2 + F_I \tag{1}$$

where the total free energy densities of the layer 1 and layer 2 are

$$F_1 = \int_{-L1}^{0} \left[\begin{array}{c} \frac{\alpha_1 (T-T_{C1})}{2} P^2 + \frac{\beta_1}{4} P^4 + \frac{\varphi_1}{2}\left(\frac{dP}{dz}\right)^2 \\ -EP\, dz + \frac{\varphi_1}{2\delta_1}(P_1^2 + P_2^2) \end{array} \right] \tag{2}$$

and

$$F_2 = \int_0^{L2} \left[\frac{\alpha_2 (T - T_{C2})}{2} Q^2 + \frac{\beta_2}{4} Q^4 + \frac{\varphi_2}{2}\left(\frac{dQ}{dz}\right)^2 - EQ \right] dz + \frac{\varphi_2}{2\delta_2}(Q_1^2 + Q_2^2) \tag{3}$$

respectively. P and Q are the order parameters (polarizations) of the layer 1 and layer 2, respectively. α_1, α_2, β_1, β_2, φ_1 and φ_2 are all positive constants. The transition temperature of the superlattice T_{CS} is defined as $T_{C1} > T_{CS} > T_{C2}$. P_1 and P_2 are the polarizations at the left boundary, $z = L_1$ and the right boundary, $z = 0$, respectively, for the FE layer. While for the PE layer, Q_1 and Q_2 are the corresponding polarizations at the left boundary, $z = 0$ and the right boundary, $z = L_1$. δ_1 and δ_2 are the extrapolation lengths for the FE layer and PE layer, respectively.

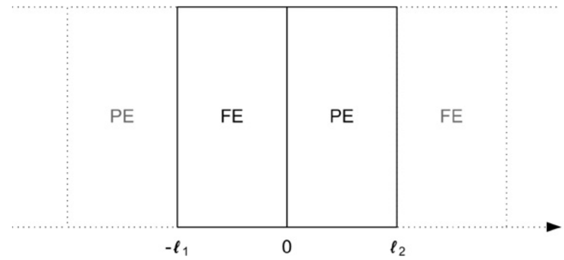

Fig. 1. Schematic illustration of a superlattice composed of a ferroelectric and paraelectric layers with the thicknesses ℓ_1 and ℓ_2, respectively.

The interlayer coupling energy of the superlattice is given by [14]

$$F_I = -J P_{av} Q_{av} \tag{4}$$

where $P_{av} = 1/L_1$ and $Q_{av} = 1/L_2$ are the average polarizations of the constituent layer 1 and layer 2, respectively. Due to the assigned negative interlayer coupling energy in this study, which is opposite to Ref. [14], $J > 0$ is the strength of the interlayer coupling, favoring the ferroelectric ordering.

The time dependence of the polarizations is given by the Landau–Khalatnikov equation as

$$\left.\begin{array}{l} \gamma_1 \dfrac{\partial P}{\partial \tau} = -\dfrac{\delta F}{\delta P} \\ \gamma_2 \dfrac{\partial Q}{\partial \tau} = -\dfrac{\delta F}{\delta Q} \end{array}\right\} \tag{5}$$

where γ_1 and γ_2 are the viscosity coefficients of the layer 1 and layer 2, respectively.

It is convenient to rescale the variables in Eq. (2) to Eq. (5) into their dimensionless forms, we use the following scaling

the free energy: $f = F/(\varphi_1 \alpha_1^3 T_{C1}^3/\beta_1^2)^{1/2}$,
the polarizations: $p = P/P_0$, $q = Q/P_0$ where $P_0 = (\alpha_1 T_{C1}/\beta_1)^{1/2}$
the Landau parameters: $\alpha_r = \alpha_2/\alpha_1$, $\beta_r = \beta_2/\beta_1$, $\varphi_r = \varphi_2/\varphi_1$
the temperatures: $t = T/T_{C1}$, $t_r = T_{C2}/T_{C1}$
the lengths: $\zeta = Z/Z_0$, $\ell_1 = L_1/z_0$, $\ell_2 = L_2/z_0$, $\delta_{r1} = \delta_1/Z_0$, $\delta_{r2} = \delta_2/Z_0$ where $Z_0 = \varphi_1/\alpha_1 T_{c1}$
the electric field: $e = E/E_0$ where $E_0 = (\alpha_1^3 T_{C1}^3/\beta_1)^{1/2}$,
and coupling constant: $j = J/(\varphi_1 \alpha_1 T_{C1})^{1/2}$.

The free energy equation of the superlattice is then reduced to dimensionless form shown below.

$$\begin{aligned} f = & \int_{-\ell_1}^{0} \left[\frac{1}{2}(t-1)p^2 + \frac{1}{4}p^4 + \frac{1}{2}\left(\frac{dp}{d\zeta}\right)^2 - ep \right] d\zeta + \frac{1}{2\delta_{r1}}(p_1^2 + p_2^2) \\ & + \int_0^{\ell_2} \left[\frac{\alpha_r}{2}(t - t_r) q^2 + \frac{\beta_r}{4} q^4 + \frac{\varphi_r}{2}\left(\frac{dq}{d\zeta}\right)^2 - eq \right] + \frac{\varphi_r}{2\delta_{r2}}(q_1^2 + q_2^2) \\ & - j p_{av} q_{av} \end{aligned} \tag{6}$$

with the boundary conditions

$$\begin{aligned} \frac{dp}{d\zeta} &= \frac{p_1}{\delta_{r1}} && \text{at } \zeta = -\ell_1 \\ \frac{dp}{d\zeta} &= -\frac{p_2}{\delta_{r1}} && \text{at } \zeta = 0 \\ \frac{dq}{d\zeta} &= \frac{q_1}{\delta_{r2}} && \text{at } \zeta = 0 \\ \frac{dq}{d\zeta} &= -\frac{q_2}{\delta_{r2}} && \text{at } \zeta = \ell_2 \end{aligned} \tag{7}$$

In dimensionless form, the Landau–Khalatnikov equations are

$$\left.\begin{aligned} \frac{\partial p}{\partial \tau_r} &= -\frac{\delta f}{\delta p} \\ \gamma_r \frac{\partial q}{\partial \tau_r} &= -\frac{\delta f}{\delta q} \end{aligned}\right\} \tag{8}$$

where $\tau_r = \tau(\varphi_1\alpha_1 T_{C1})^{1/2}/\gamma_1$ and $\gamma_r = \gamma_2/\gamma_1$.

The average polarization of the superlattice is defined as

$$\bar{p} = \frac{1}{\ell_1 + \ell_2}(+) \tag{9}$$

and the current response is obtained as

$$i = \frac{d\bar{p}}{d\tau_r}. \tag{10}$$

In the present study, the switching characteristics are obtained using the applied electric field e as

$$e = e_0 \sin(2\pi f \tau_r) \tag{11}$$

where e_0 and f denote the amplitude and frequency, respectively.

3. Results and discussion

In the calculations, the following parameters are used unless otherwise specified: $\alpha_r = 10$, $\beta_r = 10$, $\varphi_r = 0.2$, $t_r = 0.1$, $t = 0.5$, $j = 0.6$, $\delta_{r1} = 2$, $\delta_{r2} = -0.5$, $\gamma_r = 1$, $\ell_1 = \ell_2 = 3$, $e_0 = 0.2$ and $f = 0.04$. Based on the adopted parameters, the polarization is suppressed near the ferroelectric surface ($\delta_{r1} = 2$). On the other hand, the polarization is induced at the surface of the paraelectric layer ($\delta_{r2} = -0.5$), if the interlayer coupling $j \neq 0$.

We first examine the switching characteristic by applying a series of pulsed field, as shown in Fig. 2. The initial polarization is set at negative state. It is seen that the polarization changes from the initially negative to positive state when the first pulse is applied, which is characterized by the peak value i_{peak} in the current response. When the second pulse is applied, the current response is different from that of the first. This is expected because it is a non-switching current response. As the applied field is removed, the decay of polarization leads to a negative current flow, which is the well-known back-switching phenomenon. A complete switching and non-switching currents are generated by the third and fourth pulses, as shown.

We now investigate the switching characteristics of a superlattice by applying a sinusoidal field, which is generated by Eq. (11). The effect of various parameters such as field amplitude, layer thickness, interlayer coupling, temperature and viscosity coefficient on the switching characteristics are examined by looking at the $\bar{p}$–e hysteresis loop and i–e curves. In Fig. 3, we show the applied field dependence of switching characteristics, where the parameter is the field amplitude e_0. As the field amplitude e_0 increases, both the polarization and coercive field increase. The peak value of the current response i_{peak} also becomes larger with increasing e_0. The results clearly reproduce the well-known experimental results.

Fig. 4 illustrates the influence of interlayer coupling in the switching characteristics. The field amplitude is chosen as $e_0 = 1$, which is about one and a half times larger than the coercive field of the bulk ferroelectric layer at $t = 0.5$. The inset shows the polarization profiles at the initial negative state for

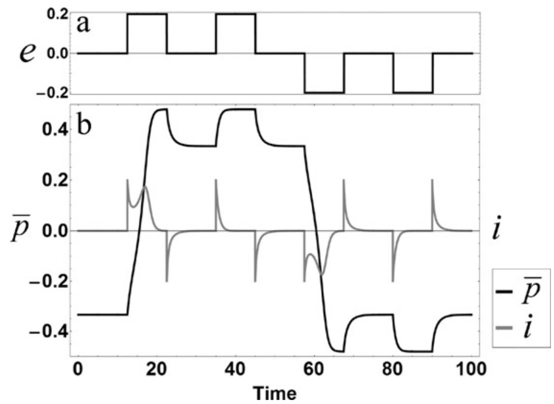

Fig. 2. Switching characteristics: (a) pulse field (b) polarization versus time and switching current versus time. The parameter adopted in the calculations are: $\alpha_r = 10$, $\beta_r = 10$, $\varphi_r = 0.2$, $t_r = 0.1$, $t = 0.5$, $j = 0.6$, $\delta_{r1} = 2$, $\delta_{r2} = -0.5$, $\gamma_r = 1$, $\ell_1 = \ell_2 = 3$, $e_0 = 0.2$ and $f = 0.04$.

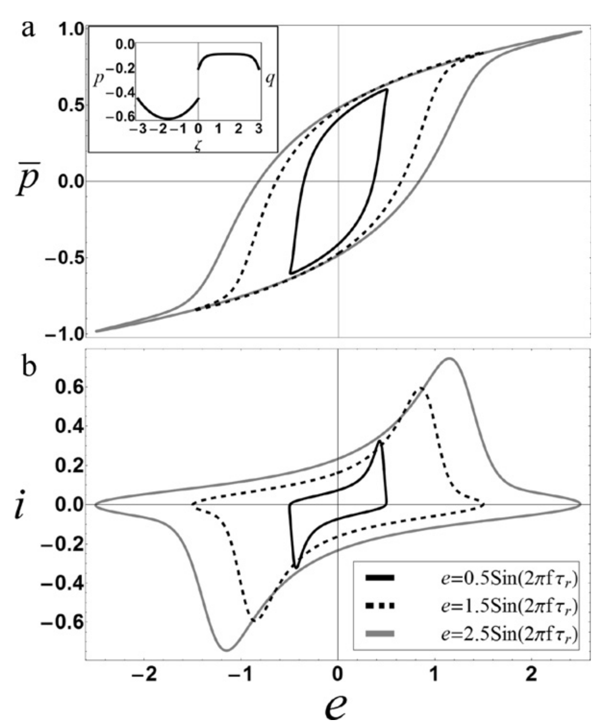

Fig. 3. Applied field e dependence of switching characteristics: (a) $\bar{p}$–e hysteresis loop, (b) i versus e. The inset shows the polarization profile. Other parameters are the same as those in Fig. 2.

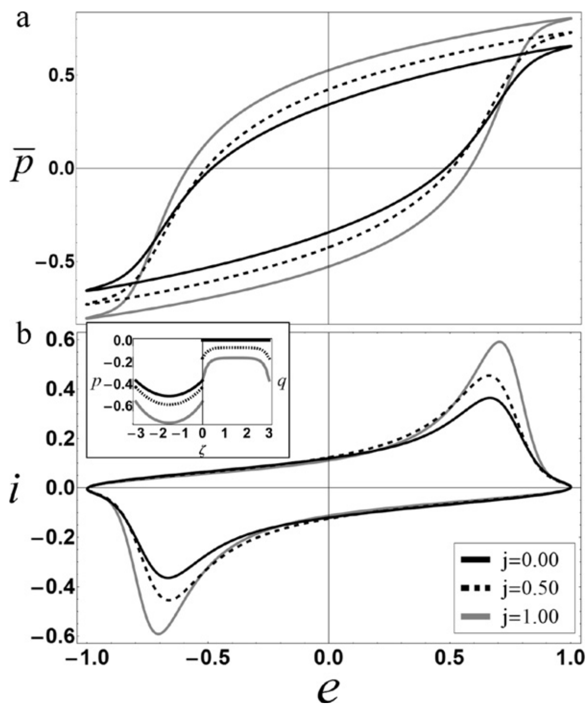

Fig. 4. Interlayer coupling dependence of switching characteristics with $e=\sin(2\pi f\tau_r)$ and $e_0=1$: (a) $\bar{p}-e$ hysteresis loop and (b) i versus e. The inset shows the polarization profiles for the three values of j interlayer coupling. Other parameters are the same as those in Fig. 2.

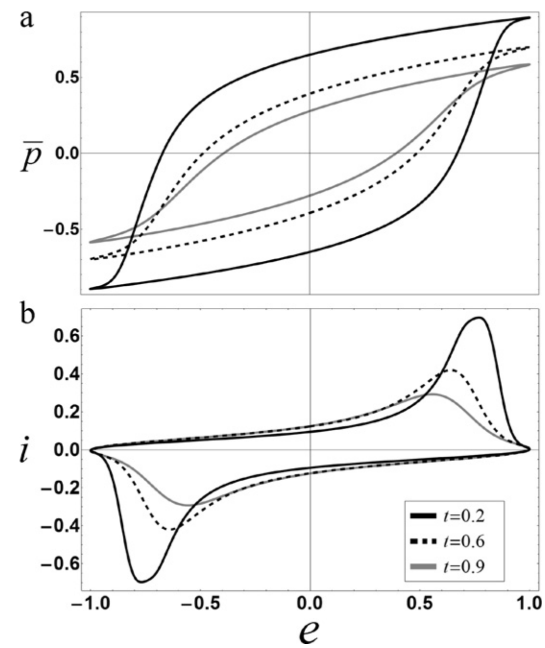

Fig. 6. Temperature dependence of switching characteristics with $e=\sin(2\pi f\tau_r)$ and $e_0=1$: (a) $\bar{p}-e$ hysteresis loop and (b) i versus e. Other parameters are the same as those in Fig. 2.

three different interlayer coupling strengths. If $j=0$, the two constituent layers are independent and no polarization is induced at the paraelectric layer. The presence of interlayer coupling $j\neq 0$ induces the polarization at the paraelectric layer, and enhances the overall polarization of the ferroelectric layer. A strong interlayer coupling leads to a significant enhancement of polarization in the superlattice. Thus, we see that the polarization, coercive field and peak current i_{peak} are enhanced with increasing j.

The effect of layer thickness on the switching characteristics of superlattice is shown in Fig. 5. As the thickness of the ferroelectric layer ℓ_1 decreases, the polarization reduces. The reduction of the polarization due to the surface effect is determined by the extrapolation length δ_r. Thus, the polariza-

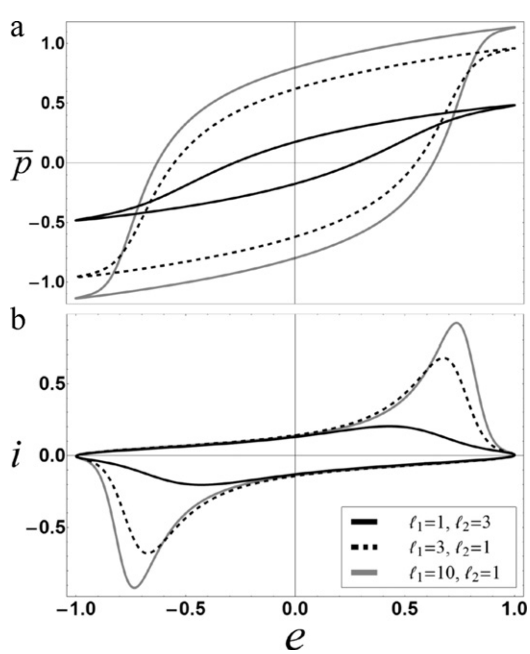

Fig. 5. Thickness dependence of switching characteristics with $e=\sin(2\pi f\tau_r)$ and $e_0=1$: (a) $\bar{p}-e$ hysteresis loop and (b) i versus e. Other parameters are the same as those in Fig. 2.

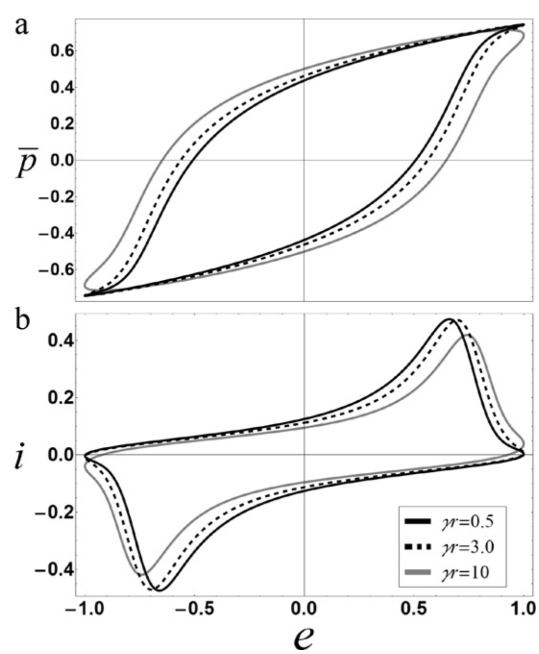

Fig. 7. Viscosity dependence of switching characteristics with $e=\sin(2\pi f\tau_r)$ and $e_0=1$: (a) $\bar{p}-e$ hysteresis loop and (b) i versus e. Other parameters are the same as those in Fig. 2.

tion, coercive field and the peak current decrease with decreasing the thickness of the ferroelectric layer ℓ_1.

In Fig. 6, we show how the temperature affects the $\bar{p}-e$ hysteresis loop and i–e curve. Our calculations show that the polarization, coercive field and peak current reduce with increasing temperature t from 0.2 to 0.9. This is expected because as the temperature approaches the phase transition temperature of the ferroelectric layer at $t = 1.0$, the polarization goes to zero. Finally, we study the influence of the viscosity on the switching characteristic of the superlattice. Fig. 7 shows the relative viscosity coefficient γ_r dependence of the $\bar{p}-e$ hysteresis loop and i–e curve. The viscosity coefficient γ_r is associated with the movement of polarization during switching. A large γ_r delays the movement of polarization. We see that both the polarization and coercive field decrease, as γ_r decreases. This suggests that the switching time is shorter for smaller coefficient γ_r.

4. Conclusion

In the present work, we have developed a model to describe a superlattice consisting of a periodic FE/PE layers within the framework of the Landau–Ginzburg theory. Switching characteristics such as the $\bar{p}-e$ hysteresis loop and i–e curve are studied using the Landau–Khalatnikov equation. Our results indicate that the field amplitude, layer thickness, interlayer coupling, viscosity, and temperature can strongly affect the switching characteristics of the superlattice. The interlayer coupling J can be related to the long-range dipole–dipole interaction within the neighbouring sublattices of the superlattice. In this study, we have used $J > 0$, which gives the ferroelectric interlayer coupling. Further calculations can be done on the combination of thickness, the sign and magnitude of interlayer coupling J to look into the thickness modulation effect and antiferroelectric behaviour in certain structures of superlattices, which were observed in experimental work [5,6,21].

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