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Adjustable ferroelectric properties in paraelectric/ferroelectric/paraelectric trilayers

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Abstract

A trilayer paraelectric/ferroelectric/paraelectric system is studied within the framework of the Ginzburg–Landau–Devonshire theory in consideration of the elastic interactions between each layer. An analytic expression of the ferroelectric phase transition temperature for the ferroelectric layer is derived. The polarization, dielectric properties and the response to the external field are studied numerically. Our results show that there are two types of thickness effects on the properties of the film considering the effect of dislocation. By changing the thickness of each layer, the ferroelectric layer can be highly adjusted and may have very good potential applications such as transducers, sensors and actuators.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Ferroelectrics have been extensively studied because of their wide and potential applications in many fields such as memory, high frequency electronics, capacitors, electrocaloric, electro-optic, piezoelectric and magnetoelectrics multifunctional devices [1–3]. Properties of the ferroelectrics can be affected by a myriad of factors, including ambient temperatures, boundary conditions, sample dimensions, misfit epitaxial stresses and defects such as dislocations and vacancies [4]. With the development of film growth, atomic-scale control of films and multilayers is expected to produce strong property enhancements and combinations of desired properties [5]. Recently, more attention has been paid to ferroelectric graded thin films, layered multiphase structures, ultrathin films, nanotubes, nanoparticles and nanowires [6–10]. Ferroelectrics with high tunability are now an issue of keen interest because of the potential application in microwave devices and sensitive functional devices such as electronic scanning antennas, tunable filters, varactors, voltage controlled oscillators, delay lines and IR detection [11]. The tunability of ferroelectrics is the dependence of the dielectric permittivity on the applied dc bias electric field with the trend 'the higher the dielectric constant, the higher the tunability' [3]. It was found that graded films are usually with larger tunability and can be obtained mainly through ways such as the graded temperature, stress and composites. However, tunable ferroelectrics, in other sense, can be gained by applying strain and stress [12] for design of the material as needed.

Multilayered epitaxial thin films are a promising solution for application in kinds of functional devices. Composites, lattices mismatch stresses and thicknesses of relative layers can greatly influence the properties of the FE layer [13]. With a proper design, we can get the desired properties. In this work, we study a trilayer system within the framework of the time-dependent Ginzburg–Landau–Devonshire (TDGLD) theory.

2. Thermodynamic model

We consider the paraelectric/ferroelectric/paraelectric (PE/FE/PE) trilayer system covered by hard electrodes as shown in figure 1, where h and H are the thicknesses of the FE and PE layers, respectively. The top and bottom PE layers have the same thickness. We set the coordinate system $z = 0$ at the middle of the PE layer, and the free surfaces at $z = h/2 + H$

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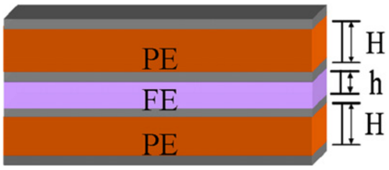

Figure 1. Schematic of the calculation model.

and $z = -(h/2 + H)$, respectively. Because of the high symmetry of the system, we assume that there is no bending in the layered structure.

2.1. *Free energy*

We use FE material with lattice parameter smaller than that of the covered PE material. In our system, we use $SrTiO_3$ as the PE material and $BaTiO_3$ as the FE material. If the lattices are well matched, the FE crystal will be highly constrained. Under the phase transition temperature, the $BaTiO_3$ layer constrained in the x–y plane will have a spontaneous polarization with the direction perpendicular to the plane, that is $P_1 = P_2 = 0$ and $P_3 = P(z)$. The total free energy per unit area of the trilayer can then be written as

$$\Delta F = F_P + F_{\mathrm{Ele}} + F_{\mathrm{Surf}} + F_{\mathrm{Ela}} = \int_{-(h/2+H)}^{h/2+H} [f_P(z) + f_{\mathrm{Ele}}(z) + f_{\mathrm{Surf}}(z) + f_{\mathrm{Ela}}(z)]\,\mathrm{d}z, \quad (1)$$

where F_P is the self-energy of the reference state which is free from the external electrical and mechanical fields. F_{Ele}, F_{Surf} and F_{Ela} are contributions due to actions on the reference state caused by electric fields, surface and mechanical stresses, respectively. f_P, f_{Ele}, f_{Surf} and f_{Ela} are the corresponding energy densities.

If the paraelectric layers do not have phase transitions, the first term of equation (1) will only be the contribution of the ferroelectric layer and can be written as the sum of the nonlinear Landau free-energy function for uniform infinite crystal and the Ginzburg contribution to account for the presence of non-uniformity in P:

$$F_P = \int_{-h/2}^{h/2} \frac{1}{2}A(T - T_{\mathrm{c0}})P^2 + \frac{1}{4}BP^4 + \frac{1}{6}CP^6 + \frac{1}{2}D_{11}\left(\frac{\mathrm{d}P}{\mathrm{d}z}\right)^2 \mathrm{d}z, \quad (2)$$

where A, B and C are the expansion coefficients of the Landau free energy associated with the dielectric stiffness and higher-order stiffness coefficients at constant stress [14], T_{c0} is the Curie–Weiss temperature of the bulk material, D_{11} is the gradient energy coefficients and can be approximated as $\xi^2 \cdot |A(T - T_{\mathrm{c0}})|$ and ξ is the characteristic length along which the polarization varies.

The second term in equation (1) is the contribution from the depolarization and external electric fields E_{d} and E_{ext}, respectively. The depolarization field can be obtained by solving the electrostatic equilibrium $\nabla \cdot D = 0$ with special boundary condition such as the short circuit or open-circuit boundary conditions [15]. With possible charge compensation, we may assume for simplicity that E_{d} is negligible. The contribution f_{Ele} can be written as

$$F_{\mathrm{Ele}} = \int_{-h/2}^{h/2} [-E_{\mathrm{ext}}P(z)]\,\mathrm{d}z. \quad (3)$$

For a material with finite size, there is a surface relaxation because of the existence of the surface. A simplified description of the surface energy can be given as follows:

$$F_{\mathrm{surf}} = \frac{D_{11}}{2}\int_s \frac{P^2}{\delta}\,\mathrm{d}s, \quad (4)$$

where s is the surface area of the element and δ is the extrapolation length along the z-axis.

The extrapolation length describes the difference between the surface and the bulk due to the surface relaxation on the polarization [16, 17]. The values can be positive or negative, corresponding to a reduction of polarization or an enhancement of polarization near the surfaces, respectively. The former is a more common case in ferroelectric materials. In general, it can be affected by boundary conditions and should be determined experimentally or by first-principle calculations for different materials [18].

The elastic energy contribution F_{Ela} in the trilayer is the interaction of the external stress and phase transition stress. Generally, the ferroelectric phase transition is caused by a change of crystallographic structures associated with a transformation strain, which is also known as spontaneous strain or eigenstrain. For materials free from any other external force and surface constraint, the eigenstrain is $\varepsilon_{ij}^P = Q_{ijkl}P_kP_l$ [19], Q_{ijkl} is the electrostrictive coefficient tensor. In this case, the polarization P is along the z direction and the transformation strain can be expressed as $\varepsilon_{11}^P = \varepsilon_{22}^P = \varepsilon^P = Q_{12}P^2$. With a coherent interface, the in-plane misfit strain in the FE layer can be written as $\varepsilon_{11}^m = \varepsilon_{22}^m = \varepsilon^m = (a_s^{\mathrm{eff}} - a_f)/a_f$. a_s^{eff} is an introduced effective substrate lattice parameter because of the relaxation of the lattice mismatch by formation of dislocations. The dislocation density varies with the thickness of the film. We introduce such thickness-dependent relaxation as the classical description [20]

$$a_{\mathrm{s}}^{\mathrm{eff}} = \frac{a_{\mathrm{s}}}{\rho a_{\mathrm{s}} + 1}, \qquad \rho = \frac{\varepsilon_0}{a_f}\left(1 - \frac{h_\rho}{h}\right), \quad (5)$$

where a_{s} is the lattice parameter of the substrate and h is the thickness of the FE layer. ρ is the equilibrium linear dislocation density at the deposition temperature, $\varepsilon_0 = (a_{\mathrm{f}} - a_{\mathrm{s}})/a_{\mathrm{s}}$ and a_{f} are the average misfit strain and in-plane lattice parameter of the film at the growth temperature, respectively. h_ρ is the average critical thickness, below which dislocation is not feasible. We should note that h_ρ will be different with different thicknesses of the substrate or the PE layer in the model [21]. Because of the limitation of the experiment data about the substrate thickness effect on the dislocation, we take h_ρ as a constant with different substrate thicknesses.

We consider both the FE and PE layers are cubic elastic media with moduli C_{11}, C_{12} and $\bar{C}_{11}$, $\bar{C}_{22}$, respectively. The effective elastic constants are given by $G = C_{11} + C_{12} - 2C_{12}^2/C_{11}$ and $\bar{G} = \bar{C}_{11} + \bar{C}_{12} - 2\bar{C}_{12}^2/\bar{C}_{11}$. Therefore, stresses in the trilayer that are caused by the combined effects of the misfit and transformation strains can be calculated by following the method of Timosheko [21]. Starting with the uncoupled FE and PE layers, the unconstrained incompatible in-plane strains are $\varepsilon_{11} = \varepsilon_{22} = \varepsilon^m - Q_{12}P^2$ in the FE layer and $\bar{\varepsilon}_{11} = \bar{\varepsilon}_{22} = 0$ in the PE layer. Uniform stresses are then imposed on each layer to achieve displacement compatibility and equilibrium, i.e. uniform strain and zero net force [12]. Finally, the constraint of strain is relaxed uniformity, the built-in or the residual equilibrated uniform stress ε can be found in the final configuration. For bilayer structures or asymmetry laminates, the stress distribution usually varies along the thickness direction and results in bending [4]. In this work, we did not consider the bending of the system because of the symmetry of the trilayer.

Taking into account both the misfit and transformation strains, the stresses σ_s and σ_f in the substrate and film are given by $\sigma_s = \bar{G}\varepsilon$ for $-H \leqslant z \leqslant -h/2$, $h/2 \leqslant z \leqslant H$ and $\sigma_f = G(\varepsilon - Q_{12}P^2 + \varepsilon^m)$ for $-h/2 \leqslant z \leqslant h/2$, respectively. The strain distribution in the trilayer system is determined by the equilibrium condition

$$\int_{-H-h/2}^{-h/2} \bar{G}\varepsilon\,\mathrm{d}z + \int_{h/2}^{H+h/2} \bar{G}\varepsilon\,\mathrm{d}z + \int_{-h/2}^{h/2} G(\varepsilon + \varepsilon^m - Q_{12}P^2)\,\mathrm{d}z = 0. \tag{6}$$

The uniform strain component ε from equation (6) can be solved as

$$\varepsilon = -\frac{Gh\left(\varepsilon^m - \frac{Q_{12}}{h}\int_0^h P^2\,\mathrm{d}z\right)}{2\bar{G}H + Gh}. \tag{7}$$

Finally, we can find the total elastic energy F_{Ela} of the trilayer

$$F_{\mathrm{Ela}} = 2\int_{h/2}^{H+h/2} \bar{G}\varepsilon(z)\varepsilon(z)\,\mathrm{d}z + 2\int_0^{h/2} G(\varepsilon + \varepsilon^m - Q_{12}P^2)(\varepsilon + \varepsilon^m - Q_{12}P^2)\,\mathrm{d}z. \tag{8}$$

2.2. *Time-dependent Ginzburg–Landau–Devonshire equation*

The temporal evolution of the polarization field in the ferroelectric layer can be described by the TDGLD equation

$$\frac{\partial P(z,h,H,t)}{\partial t} = -M\frac{\delta F(E_{\mathrm{ext}})}{\delta P} = -M\left(A^*P + B^*P^3 + CP^5 - D_{11}\frac{d^2P}{dz^2} - E_{\mathrm{ext}}\right), \tag{9}$$

where M is the kinetic coefficient related to the domain wall mobility. With the coupling effect of the mechanical interaction between layers, the Landau expansion coefficients can be renormalized as

$$A^* = A(T - T_{c0}) - 4GQ_{12}(\varepsilon + \varepsilon^m),\qquad B^* = B + 4GQ_{12}^2. \tag{10}$$

The surface item yields the boundary condition as

$$\frac{\partial P}{\partial z} = \mp\frac{P}{\delta} \qquad \text{for } z = \pm\frac{h}{2}. \tag{11}$$

When the surface effect is negligible, that is $\delta \to \infty$, the mechanical boundary conditions for the FE layer are average strain controlled by the upper and lower PE layers.

With the boundary condition equation (11), equation (9) can be solved numerically to yield the steady-state polarization distribution. The average polarization $\langle P\rangle$ can be described as $\langle P\rangle = \frac{1}{h}\int_0^h P\,\mathrm{d}z$ and the uniform strain component ε can then be determined from equation (7).

2.3. *Linear analysis and phase transition temperature*

The appearance of spontaneous polarization is highly temperature dependent. Because of the electrostrictive properties, ferroelectric crystals undergo phase transitions always involve structural changes that will have interactions with the external stress. On the other hand, the external stress can also greatly affect properties of ferroelectric materials such as the phase transition temperatures and dielectric properties.

According to linear analysis theory, we can analyse the critical condition of the dynamic stability of the stationary state to find the phase transition temperature [22]. The dynamic stability of the stationary state can be probed by applying infinitesimal perturbation Δ to the trivial stationary solution $P_0 = 0$. Neglecting the higher order small qualities and only retaining the terms linear in Δ, equation (8) becomes

$$\frac{\partial\Delta}{\partial t} = -M\left(A^*\Delta - D_{11}\frac{\partial^2\Delta}{\partial z^2}\right). \tag{12}$$

In the same way, the boundary condition (equation (11)) can be derived with P replaced by Δ, that is, $\partial\Delta/\partial z = \mp\Delta/\delta$. The parameter δ gives different types of the surface effects. For simplicity, we consider the more common case$\delta > 0$, and the comprehensive discussion was given by Wang and Woo [15]. By separating variables and applying the boundary condition, the solution for equation (12) can be separated into time dependent and time independent parts connected by the eigenvalue:

$$\Delta(x,z,t) = \mathrm{e}^{\omega t}\varphi_\omega(x,z) = \Delta_0\mathrm{e}^{\omega t}\cos(kz), \tag{13}$$

where ω is the eigenvalue and φ_ω is the corresponding eigenfunction. According to the stability analysis, the critical condition is $\omega = 0$. Thus the supercooling temperature is

$$T_c = T_{c0} - \frac{1}{A}(D_{11}k_c^2) + \frac{1}{A}4GQ_{12}(\varepsilon + \varepsilon^m). \tag{14}$$

Substituting equation (13) into the boundary condition, k_c can be found as the smallest values of k. Substituting k_c into equation (14), we can obtain the critical phase transition temperature which is highly dependent on the external stress.

2.4. *Tunable properties of the FE layer*

As the temperature decreases from a high temperature that is above the Curie temperature, a structural phase change takes place and the crystal changes from paraelectric to ferroelectric accompanied with a large increase in the dielectric constant. By changing the phase transition temperature, we can tune the dielectric constant to proper values in a given ambient temperature.

For layered dielectric thin films, the dielectric permittivity and the tunability can be strongly affected by the 'passive' layer. The system can be described as the in-series connection of several capacitors. Electrically, the effective dielectric permittivity of such composite can be written as [3] $\frac{1}{\varepsilon_{\mathrm{mix}}} = \frac{1-q}{\varepsilon_{\mathrm{P}}} + \frac{q}{\varepsilon_{\mathrm{F}}}$, where q is the volume concentration of the ferroelectric phase. $\varepsilon_{\mathrm{mix}}$, ε_{P} and ε_{F} are the dielectric constants of the composite, ferroelectric and paraelectric, respectively.

In this paper, we focus only on the properties of the FE layer. The dielectric susceptibility of the FE layer can be determined as the small-signal dielectric response $\chi_{\mathrm{F}} = \left(\varepsilon_0 \frac{\partial^2 f}{\partial P^2}\right)^{-1}\Big|_{E_{\mathrm{ext}}=0}$, the form of which is given by $\chi_{\mathrm{F}}^{-1} = \varepsilon_0(A^* + 3B^*\langle P\rangle^2 + 5C\langle P\rangle^4)$ with $\langle P\rangle$ solved from the static state in the absence of external field. ε_0 is the permittivity of free space.

2.5. *Response of the trilayer under a cyclic electric field*

It is well known that the properties of ferroelectrics can be greatly affected by the external stress. Different constraints or tensions can lead to different material responses to the external field. An in-plane constraint can produce a large enhancement of the remnant polarization and the coercive field. We simulated the hysteresis loop by solving the TDGLD equation which governs the time evolution of the FE layer under a cyclic sinusoidal external field E_{ext} along the z direction:

$$E_{\mathrm{ext}} = E_0 \sin\left(\frac{2\pi t}{T_{\mathrm{E}}}\right) = E_0 \sin(2\pi t f_{\mathrm{E}}), \qquad (15)$$

where E_0, T_{E} and f_{E} are the amplitude, period and frequency of the sinusoidal electric field, respectively.

3. Results and discussions

We studied a specific example of the $SrTiO_3/BaTiO_3/SrTiO_3$ trilayer system. The material constants for the Landau free energy, the electrostrictive coefficients and the elastic properties are from [6, 23]. For simplicity, we take the two PE layers with the same thickness. The average polarization $\langle P\rangle$ of the FE layer is calculated by solving equation (9) without external field. With the thickness-dependent dislocation effect, the spontaneous polarization has a different trend with the thickness of the PE layer as shown in figure 2(*a*). When the FE layer is rather thin, the average polarization P increases with increasing thicknesses of the PE layers; while the FE layer is thicker than about 100 nm, P decreases with increasing thicknesses of the PE layers. The reasons for the two cases are as follows. (i) When the FE layer is rather thin, there are a few dislocations during deposition and the misfit strain with a little relaxation is larger than the spontaneous eigenstrain at the stable state, that is $|Q_{12}\langle P\rangle^2| \leqslant |\varepsilon^m|$, which results in a tensile residual strain. According to equation (7), the tensile stain decreases with increasing PE layer thickness, resulting in an enhancement of the spontaneous polarization and an increase in the phase transition temperature. We should note that the thickness of the FE layer we considered is larger than the critical dislocation thickness h_ρ and the thickness effect is mainly due to the dislocation relaxation. For ultrathin film whose thickness is less than 30 nm, surface effect, leakage current, occurrence of the dead layer and the depolarization field will be very significant and usually the spontaneous polarization will decrease with the decrease in the film thickness [9, 24, 25]. (ii) When the FE layer is thick, the misfit strain is relaxed and smaller than the spontaneous strain, that is $|Q_{12}\langle P\rangle^2| \geqslant |\varepsilon^m|$, which results in a compressive residual strain. In this case, the PE layer is a 'passive' layer to the FE layer. With the increase in the PE layer thickness, the spontaneous polarization and the phase transition temperature are deduced (figure 2).

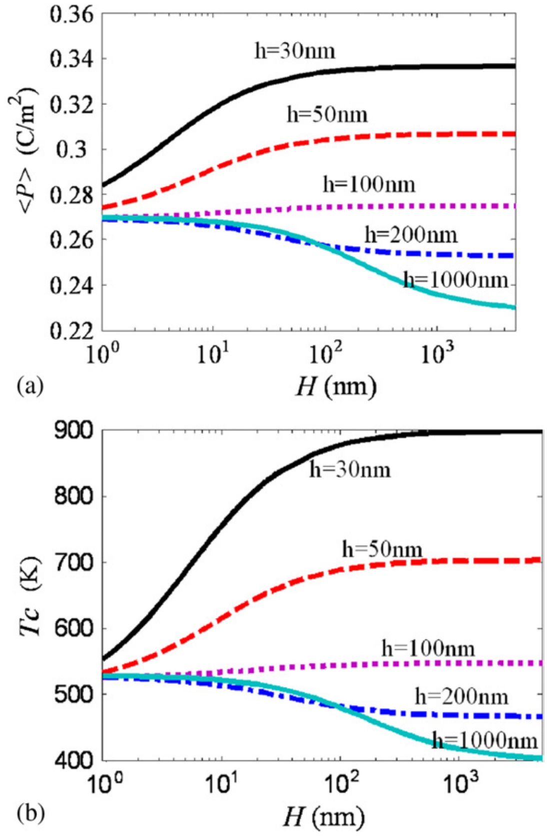

Figure 2. (*a*) Average polarization and (*b*) phase transition temperature of the FE layer with different thicknesses versus the thicknesses of the PE layers.

The dielectric susceptibility versus temperature with different ratios of layer thickness is given in figure 3, where the thickness of the FE layer is 30 nm. The cases when the FE layer is thin or thick are different as expected associated with the spontaneous polarizations and phase temperatures. By adjusting the thickness of each layer, large dielectric susceptibilities can exist in a broad area of temperature.

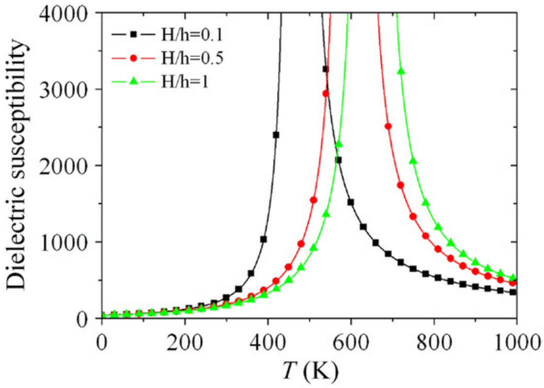

Figure 3. Dielectric susceptibility versus temperature with different ratios of layer thickness.

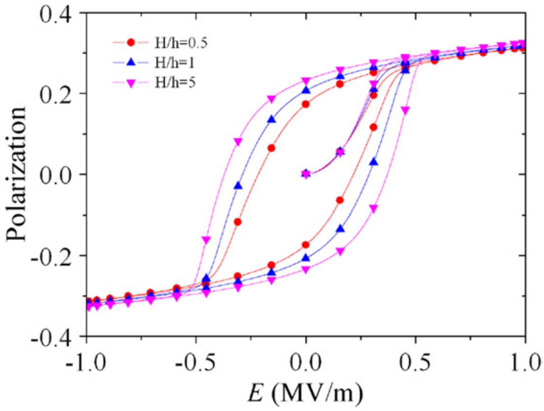

Figure 4. Response of the system to the external field with different ratios of layer thickness.

The response of the system to the external field is also simulated as shown in figure 4. We compare the cases where $H/h = 0.1, 1$ and 5 with $h = 30$ nm. The average polarization $\langle P\rangle$ is calculated as a function of a sinusoidal applied electric field by solving equation (9). In equation (15), the amplitude of the external field used is $E_0 = 100\,\mathrm{MV\,m^{-1}}$. From the hysteresis loops of the trilayer, we can see that the remnant polarization P_r and coercive field E_c are enhanced as H increases when the FE layer is thin and weakened when the FE layer is thick.

4. Conclusions

In summary, an elastic and thermodynamic model is constructed for investigating the properties of a PE/FE/PE trilayer system within the framework of Ginzburg–Landau–Devonshire theory. Considering the effects of the dislocations and misfit strain between the layers, we obtained static and dynamic polarization, phase transition temperatures and dielectric properties as a function of the layer thicknesses. Our results show that the properties of the FE layer are greatly affected by the PE layer and can be adjusted by varying the thicknesses of the relative layers. The elastic and thermodynamic model developed in this paper is applicable to many other multilayer systems.

Acknowledgments

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