The hysteresis loops of a ferroelectric bilayer film with surface transition layers

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Based on the transverse Ising model in the framework of the mean field approximation, this paper discusses a ferroelectric bilayer film with the surface transition layers within each constituent slab and an antiferroelectric interfacial coupling between two slabs. The hysteresis loop of a bilayer film is investigated. The results show that the surface transition layer in a ferroelectric bilayer film plays a significant role in realizing the multiple-state memory.

Keywords: ferroelectric bilayer film, transverse Ising model, hysteresis loops, surface transition layers
PACC: 7755, 7730, 7780B

1. Introduction

Ferroelectric bilayer films or superlattice structures and multilayers have been extensively discussed in recent years due to the rapid development of experimental techniques which make it possible to perceive well characterized samples.[1−8] These materials exhibit some excellent properties, such as an enhancement in the remnant polarisation,[9,10] extremely high dielectric response,[11] lower losses and high electric breakdown field,[12] multiple hysteresis loops,[13] and so on, which are not achieved either in the bulk or in the solid solution. Concerning these phenomena, researchers consider that the interfacial coupling between two ferroelectric materials may play a nontrivial role. Hence, there have been various studies on the properties (including polarisation, phase transition temperature and dielectric susceptibility, etc) of ferroelectric bilayer films or superlattices with the ferroelectric or antiferroelectric interfacial coupling.[14−19]

Recently, the study on the hysteresis loop of ferroelectric bilayer films or superlattices turns out to be a hotspot in both experiments and theories, because the hysteresis behaviours relate to their applications in ferroelectric memories. Experimentally, Ranjith *et al.*[20] observed that the $Pb(Mg_{1/3}Nb_{2/3})O_3$(PMN)–$PbTiO_3$ (PT) bilayer films deposited through pulsed laser ablation deposition exhibited a slim hysteresis loop with the total thickness of a bilayer film of 10 nm, and a slim double hysteresis loop with the total thickness of a bilayer film at the range of 20–50 nm. Thereafter, they observed an antiferroelectriclike polarisation hysteresis in compositionally varying multilayers of $(1-x)$ PMN–(x) PT. Triple hysteresis loops under different temperatures were clearly shown.[13] Wu *et al.*[21] studied a $(Pb_{0.90}La_{0.10})Ti_{0.975}O_3$ (PLT) / $Pb(Zr_{0.20}Ti_{0.80})O_3$(PZT) bilayer thin film fabricated by using radio frequency magnetron sputtering with a PbO_x buffer layer. Double hysteresis loops observed in the studies provided clear evidence of antiferroelectricity in PLT/ PZT bilayer films. They thought that the double loop behaviour should be attributed to the competition between the intrinsic ferroelectric coupling in the ferroelectrics and the antiferroelectric coupling among the dipoles at the interface. In theory, Ginzburg–Landau–Devonshire (GLD) phenomenological theory[22,23] as well as transverse Ising model (TIM)[24−26] with the mean-field or the effective field approximations is the major approaches to describe the above behaviours. Using the GLD theory, Chew *et al.*[22] presented that a ferroelectric bilayer or superlattice structure with antiferroelectric coupling showed interesting hysteresis loops. Their work suggested the possibility of designing multilayer elements for computer memories with multi-state memory. Based on

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the idea of Chew *et al.* the hysteresis loops of a ferroelectric bilayer film have been illustrated by considering the spatial variation of polarisation and the surface effect.[23] Using the TIM within the framework of mean field approximation, Wu *et al.*[24] investigated the hysteresis loop of a ferroelectric bilayer film with an antiferroelectric interfacial coupling. They indicated that for realizing a multiple-state memory in experiments, the thickness, interfacial coupling, quantum effect and temperature must be considered. Saber and co-workers[25,26] also studied the hysteresis loop of a ferroelectric bilayer film with an antiferroelectric interfacial coupling by means of the TIM with the effective field theory, and obtained the same results as Wu's.

As we know, Landau phenomenological theory is a macroscopic theory, thus, the quantum effect cannot be taken into consideration. The microscopic pseudo-spin theory based on the TIM can consider the quantum effect and better describe a bilayer film. However, in the above TIM theoretical studies, the properties of surface and interior of each constituent slab are assumed uniform, this assumption is not realistic because the properties of the surface are different from those of the interior for a finite size slab and should continuously change to the interior. Therefore, the surface transition layer should exist within each constituent slab. The surface transition layer of the TIM is introduced by Sun *et al.*[27] to study the effect of the gradual transition of structure on the properties of a ferroelectric thin film. It is pointed out that the effect of surface transition layer should be taken into account for a ferroelectric bilayer film or superlattice design. Thereby, using the TIM, we design a ferroelectric bilayer film to illustrate hysteresis behaviours, considering the surface transition layer in each slab and antiferroelectric interfacial coupling between the two constituent slabs. To our knowledge, the combined influence of the surface transition layer and antiferroelectric interfacial coupling on the hysteresis loop of a ferroelectric bilayer film has not been reported.

In this paper, the hysteresis loop of a ferroelectric bilayer film with an antiferroelectric interfacial coupling and the surface transition layer is presented using the TIM within the mean field approximation.

2. The model

To design such ferroelectric bilayer films or superlattice structures and multilayers, Ranjith *et al.*[20] fabricated a PT–PMN superlattice structure deposited through pulsed laser ablation deposition with different periodicities. And for a given period, the PT and PMN were of equal thickness. It is observed that when the periodicity of the PT–PMN superlattice structure is within 20–50 nm, the interfacial coupling is antiferroelectric coupling.

So we design a bilayer film consisting of two different ferroelectric slabs A and B of layer thicknesses $N_{\rm A}$ and $N_{\rm B}$, and with the surface transition layers and an antiferroelectric coupling ($J_{\rm ab} < 0$) (see Fig. 1). Each pseudo-spin layer is parallel to the $x-y$ plane with infinite size and the pseudo-spins site at square lattice points. The z-direction is perpendicular to the surface and the polarisation is along the z-direction.

The Hamiltonian of the system is described by

$$H = -\sum_i \Omega_i S_i^x - \frac{1}{2}\sum_{ij} J_{ij} S_i^z S_j^z - 2\mu E \sum_i S_i^z, \quad (1)$$

where Ω_i is the transverse field (for H-bond ferroelectrics it represents the proton tunneling between the two equilibrium positions on the H bonds) acting on the spin S_i^x at site i, S_i^x and S_i^z are the x and z components of a spin $\frac{1}{2}$operator at site i respectively (the thermal average of S_i^z is related to the average polarisation), J_{ij} is the exchange interaction constant between two-spins at site i and site j, and the sum $\sum\limits_{ij}$ only runs over the nearest-neighbour pairs, E is the applied electric field, and μ is the dipole moment on site i.

In our model, as shown in Fig. 1, we assumed that $J_{ij} = J_{\rm a}\,(m)$ ($J_{ij} = J_{\rm b}\,(m)$) if sites i and j are within the same pseudo-spin layer in A(B) slab; $J_{ij} = J_{\rm ae}\,(m)$ ($J_{ij} = J_{\rm be}\,(m)$) if two sites are across two different pseudo-spin layers in slab A(B). But, when both the two sites are inside the perfect structure layer in A(B), the exchange interaction J_{ij} is taken to be the same value as the bulk constant $J_{\rm A}$ ($J_{\rm B}$). If the two pseudo-spins site the A and B slab, respectively, in this way, $J_{ij} = J_{\rm ab}$. In this paper, we focus our attention on the case of antiferroelectric coupling ($J_{\rm ab} < 0$). We also allow Ω_i to be different between the surface and inside layer in each slab, i.e. $\Omega_i = \Omega_{\rm as}(\Omega_i = \Omega_{\rm bs})$ for site i on the surface transition layer in A(B); and for inside the perfect structure layer in A(B), Ω_i turns into $\Omega_{\rm A}$ ($\Omega_{\rm B}$) (see Eqs. (2)–(7)).

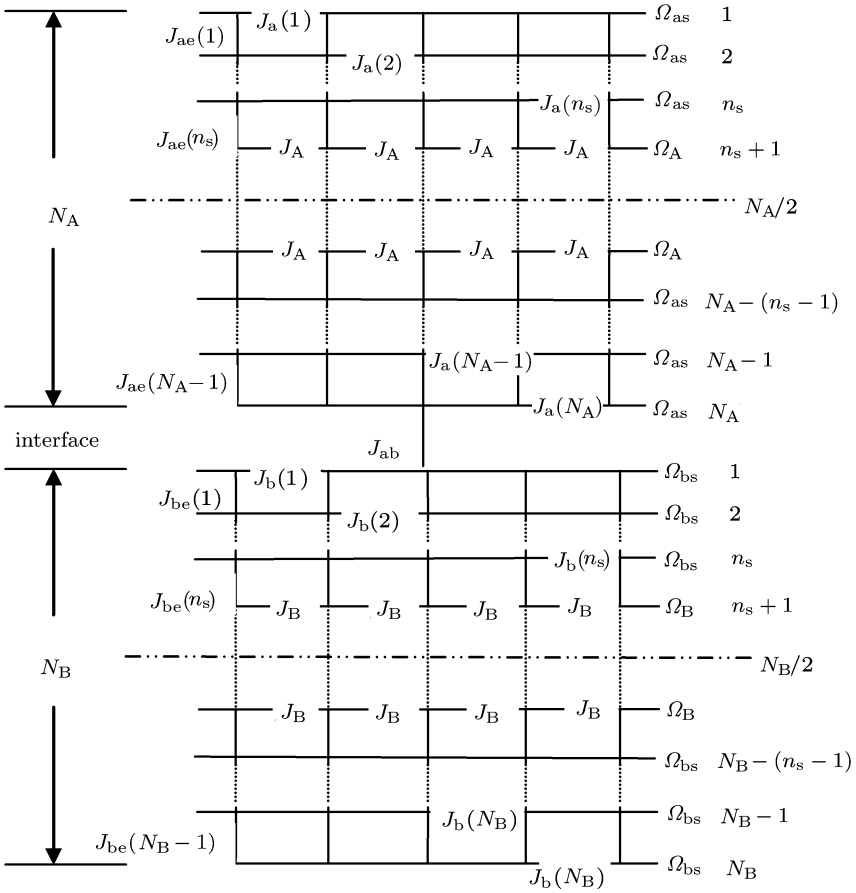


Fig. 1. Geometric structure of a bilayer film under study.

Since we do not have any experimentally measured data available on the surface imperfection, the simple functions are chosen to describe the inhomogeneous distribution of the exchange interactions and transverse fields. These particular choices do not affect the generality of the results and conclusions.

$$J_{\rm a}(m)=\alpha_1\left(\frac{m}{n_{\rm s}+1}\right)^{\sigma_1}J_{\rm A},\quad m=1\sim n_{\rm s}\ \text{and}\ m=N_{\rm A}-(n_{\rm s}-1)\sim N_{\rm A}\left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right),\tag{2a}$$

$$J_{\rm a}(m)=J_{\rm A},\quad n_{\rm s}<m<N_{\rm A}-(n_{\rm s}-1),\quad \left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right)\tag{2b}$$

$$J_{\rm ae}(m)=\alpha_2\left(\frac{m}{n_{\rm s}+1}\right)^{\sigma_1}J_{\rm A},\quad m=1\sim n_{\rm s}\ \text{and}\ m=N_{\rm A}-(n_{\rm s}-1)\sim N_{\rm A}\left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right),\tag{3a}$$

$$J_{\rm ae}(m)=J_{\rm A},\quad n_{\rm s}<m<N_{\rm A}-(n_{\rm s}-1),\quad \left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right),\tag{3b}$$

$$\Omega_{\rm as}(m)=\alpha_3\left(\frac{m}{n_{\rm s}+1}\right)^{-\sigma_1}\Omega_{\rm A},\quad m=1\sim n_{\rm s}\ \text{and}\ m=N_{\rm A}-(n_{\rm s}-1)\sim N_{\rm A}\left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right),\tag{4a}$$

$$\Omega_{\rm as}(m)=\Omega_{\rm A},\quad n_{\rm s}<m<N_{\rm A}-(n_{\rm s}-1),\quad \left(n_{\rm s}\le\frac{N_{\rm A}}{2}\right),\tag{4b}$$

$$J_{\rm b}(m)=\beta_1\left(\frac{m}{n_{\rm s}+1}\right)^{\sigma_2}J_{\rm B},\quad m=1\sim n_{\rm s}\ \text{and}\ m=N_{\rm B}-(n_{\rm s}-1)\sim N_{\rm B}\left(n_{\rm s}\le\frac{N_{\rm B}}{2}\right),\tag{5a}$$

$$J_{\rm b}(m)=J_{\rm B},\quad n_{\rm s}<m<N_{\rm B}-(n_{\rm s}-1),\quad \left(n_{\rm s}\le\frac{N_{\rm B}}{2}\right),\tag{5b}$$

$$J_{\rm be}(m) = \beta_2 \left(\frac{m}{n_{\rm s}+1}\right)^{\sigma_2} J_{\rm B}, \quad m = 1 \sim n_{\rm s} \text{ and } m = N_{\rm B} - (n_{\rm s}-1) \sim N_{\rm B}\left(n_{\rm s} \le \frac{N_{\rm B}}{2}\right), \tag{6a}$$

$$J_{\rm be}(m) = J_{\rm B}, \quad n_{\rm s} < m < N_{\rm B} - (n_{\rm s}-1), \quad \left(n_{\rm s} \le \frac{N_{\rm B}}{2}\right), \tag{6b}$$

$$\Omega_{\rm bs}(m) = \beta_3 \left(\frac{m}{n_{\rm s}+1}\right)^{-\sigma_2} \Omega_{\rm B}, \quad m = 1 \sim n_{\rm s} \text{ and } m = N_{\rm B} - (n_{\rm s}-1) \sim N_{\rm B}\left(n_{\rm s} \le \frac{N_{\rm B}}{2}\right), \tag{7a}$$

$$\Omega_{\rm bs}(m) = \Omega_{\rm B}, \quad n_{\rm s} < m < N_{\rm B} - (n_{\rm s}-1), \quad \left(n_{\rm s} \le \frac{N_{\rm B}}{2}\right), \tag{7b}$$

where m is the sequence number of the pseudo-spin layer in each slab, $N_{\rm A}$ ($N_{\rm B}$) is the total layer number across slab A(B), $n_{\rm s}$ is the number of pseudo-spin layers included in a surface transition layer in each slab. The parameter $\sigma_1(\sigma_2)$ reflects the variable intensity of the interactions (intra-layer and inter-layer) and transverse field near the surface in slab A(B). The parameters α_1, α_2, α_3 (β_1, β_2, β_3) are adjustable parameters representing the strength of the interactions (intra-layer and inter-layer) and transverse field of slab A(B).

Using the mean field approximation and assuming that the pseudo-spins in the same layer have the same values, we can express the average spin along the z direction in the ith layer[24]

$$R_m = \frac{\langle H_m^z \rangle}{2|H_m|} \tanh\left(H_m/2k_{\rm B}T\right), \tag{8}$$

where

$$\langle H_m^z \rangle = 4J_m R_m + J_{m+1}R_{m+1} + J_{m-1}R_{m-1} + 2\mu E, \tag{9}$$

$$|H_m| = \sqrt{\Omega_m^2 + \left(\langle H_m^z \rangle\right)^2}, \tag{10}$$

and m runs over each layer within the two slabs.

Equation (8) represents a set of simultaneous equations from which R_m can be calculated iteratively. The polarisation of the mth layer is proportional to the thermal average of R_m, i.e.

$$P_m = 2n\mu R_m, \tag{11}$$

where n is the number of pseudo-spins per unit volume.

Then the mean polarisation $\bar{P}$ of the bilayer is determined by

$$\bar{P} = \frac{1}{N}\sum_{m=1}^{N} P_m = 2n\mu \frac{1}{N}\sum_{m=1}^{N} R_m, \tag{12}$$

where N is the total thickness of the bilayer, i.e. $N = N_{\rm A} + N_{\rm B}$.

For simplicity, the surface transition layers in each slab are assumed symmetric, and the numbers of surface transition layers for two slabs are taken the same.

3. Numerical results and discussions

In our calculations, the parameters are selected as: $N_{\rm A} = N_{\rm B} = 6$, $J_{\rm A}/J_{\rm B} = 0.5$, $\Omega_{\rm A}/J_{\rm B} = 0.5$, $\Omega_{\rm B}/J_{\rm B} = 1/3$. We set $t = T/T_{\rm BC}$, where $T_{\rm BC}$ is the transition temperature of slab B.

A series of hysteresis loops for various values of antiferroelectric coupling constant is illustrated in Fig. 2 with the fixed parameters of surface transition layer. As shown in Fig. 2(a), if the absolute value of the antiferroelectric coupling constant $|J_{\rm ab}|$ is small, only a central loop opens. The central loop involves two steps due to the different ferroelectric properties of two constituent slabs. And the step inclines, owing to the slow increase of the applied electric field. When $|J_{\rm ab}|$ increases, the hysteresis curve changes from one central loop to triple loops. The larger the antiferroelectric coupling constant is, the more difficult for the applied electric field to change the direction of polarisation at two interfacial layers in slab A and slab B is. So the outer loops stretch further out horizontally with increasing $|J_{\rm ab}|$. The above phenomena were widely confirmed, e.g., Ref. [24] and [25]. It is implied that the behaviours of the system are ferroelectriclike for smaller $|J_{\rm ab}|$, and antiferroelectriclike for larger $|J_{\rm ab}|$. The following discussions focus on the effect of the surface transition layer on the hysteresis loops with fixed value of $J_{\rm ab}$. Synchronously, the combined influence of the surface transition layer and $J_{\rm ab}$ on the hysteresis loops of a bilayer film is elucidated in detail.

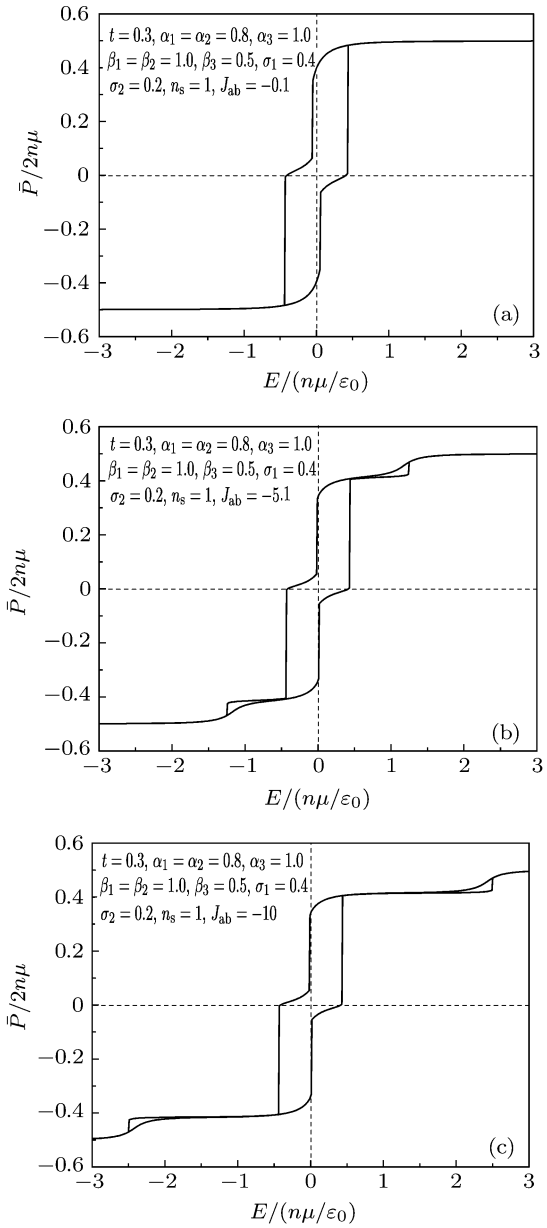


Fig. 2. The hysteresis loops of a bilayer film with different antiferroelectric interfacial coupling constants.

The effect of the σ_1 and σ_2 on the hysteresis loops is shown in Fig. 3. From Fig. 3, we can see that with increasing σ_1 or σ_2, the zone of the outer loops shrinks. In the central loop, the width of the steps clearly changes horizontally as σ_2 augments. According to Eqs. (2)–(7), when the value of the σ_1 or σ_2 is larger, the difference of physical properties between the surface transition layer and interior in slab A or slab B is greater and the polarisation of the surface transition layer becomes smaller. So with increasing σ_1 or σ_2, it is more easy to align the pseudo-spins in zones near the interface in A and B slabs along the direction of the applied electric field, and the system is ferroelectriclike. Moreover, the action of σ_2 on the hysteresis loop is more obvious than that of σ_1, because the interactions between the pseudo-spins in bulky B are stronger than those in bulky A. We further increase σ_2, as shown in Fig. 3(d). It is clearly seen that the size of the outer loops is very small, the width of the steps remarkably changes horizontally, and four steps appear. In order to analyse the mechanism of hysteresis loop, we label the directions of the pseudo-spins of surface transition layers and interior of two slabs (from slab A (left) to slab B (right)) during the course of increasing the applied electric field. We assume that the curve begins at point M. The electric field favours the layer polarisation to be aligned with the electric field, whereas the antiferroelectric interfacial coupling favours the two interfacial layer polarisations to be opposite. For the area MN, the electric field is large and has stronger influence on the layer polarisation, so the polarisations of two slabs are switched down. Point N is the start of polarisation reversal, here, the electric field decreases and the influence of the antiferroelectric coupling begins to dominate over the electric field. Due to the interactions between the pseudo-spins in slab A are weaker, thus, the interfacial layer polarisation in slab A switches up and those of the remainder of the bilayer stay down in the section $N'O$. Next switching occurs at point O. Here the electric field is positive but small in magnitude, and the influence of the antiferroelectric coupling is still stronger. Therefore, all layer polarisations of slab A switch up and those of slab B keep down for the section $P'P$. Another switching takes place at point P. The electric field further increases, all layer polarisations of two slabs switch up except the interfacial layer of slab B in the region $Q'Q$. It is implied that in the region $Q'Q$, the effect of the antiferroelectric coupling on the interfacial layer of slab B is stronger than that of the electric field. Due to the Coulomb long-range interactions, the influence of surface transition layer on the polarisation will be felt at a certain depth of a slab. So when the surface transition layer effect augments, the weaker electric field is needed to change the direction of interior layers polarisations in slab B, in comparison with the region of $P'P$ in Figs. 3(c) and 3(d). We can also conclude that if the surface transition layer effect is strong enough, the impact of the electric field on the interfacial layer of slab B will be larger than that of the antiferroelectric coupling, so that all layer polarisations will switch up. It means that the switching at point P disappears. Therefore, there are two critical values σ_{2c_1} and σ_{2c_2} in slab B.

When $\sigma_{2c_1} < \sigma_2 < \sigma_{2c_2}$, the flip of point P occurs, otherwise, it will vanish. This phenomenon has not been investigated.[24−26] In region $R'R$, the layer polarisations of slab B switch up and the interfacial layer polarisation in slab A switches down. Until point S, the electric field is large enough and positive, dominating the antiferroelectric coupling, so that all layer polarisations of two slabs switch up. The existence of the surface transition layer makes the system have ferroelectriclike characters, whereas the antiferroelectric coupling favours the antiferroelectriclike characters. Hence, the antiferroelectric coupling is an advantage of being multiple-state hysteresis.

In Fig. 4, we show the dependence of the hysteresis loops on the various values of α_1 and α_2. One can see that the central loop is almost unchanged with increasing α_1 or α_2. With increasing α_2, the size of outer loops enlarges. The reason is that the increase of the inter-layer interactions in slab A makes the electric field change hardly the direction of polarisation near the interfacial layer, which is an advantageous to the effect of antiferroelectric coupling on the layer polarisations.

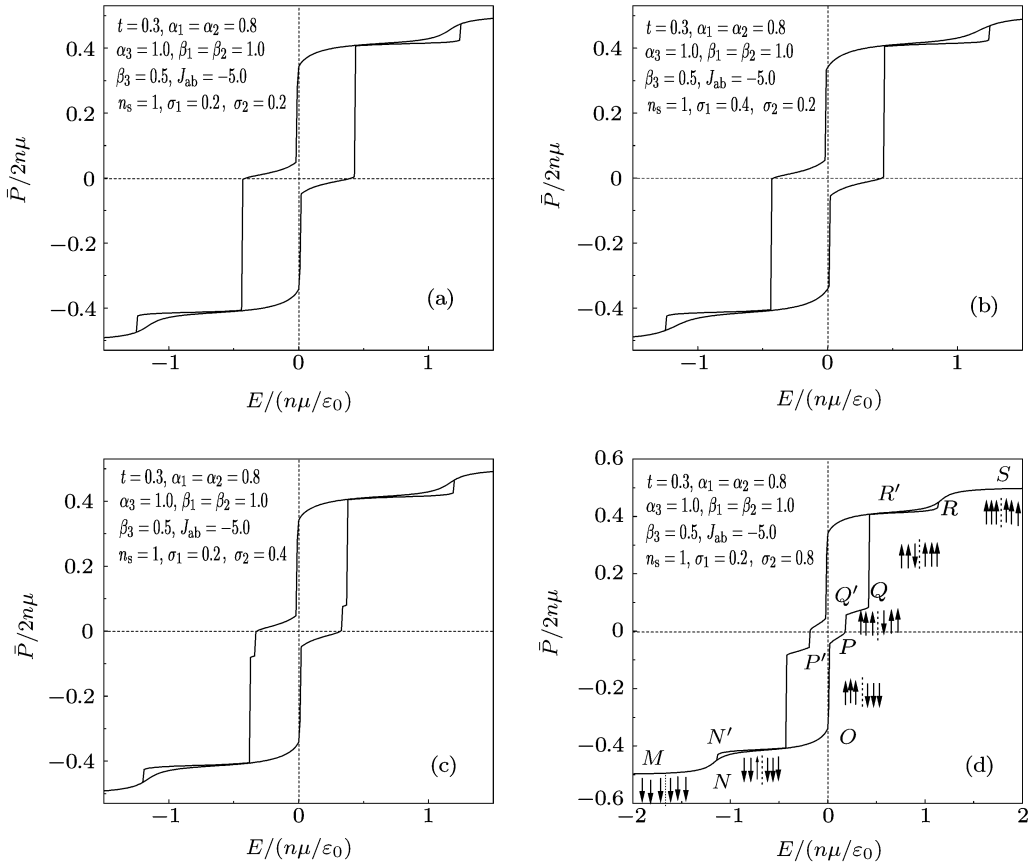


Fig. 3. The hysteresis loops patterns for various values of σ_1 and σ_2. The arrows represent the upper surface, the interior and lower surface of each slab (from slab A (left) to slab B (right)), respectively.

The hysteresis loops of a bilayer film with different values of β_1 and β_2 are shown in Fig. 5. As can be clearly seen that with increasing β_1, the size of the outer loops becomes large. With increasing β_1 or β_2, the width of the steps obviously changes horizontally. And the influence of β_1 on the hysteresis loops is more remarkable than that of β_2, i.e. the influence of the intra-layer interactions on the hysteresis loops is more evident. In addition, the four steps in the central loop appear. The reason of above phenomena is the same as that of Fig. 3. Furthermore, β_1 and β_2 have two pairs of critical values. Between the two pairs of critical values, there are four steps.

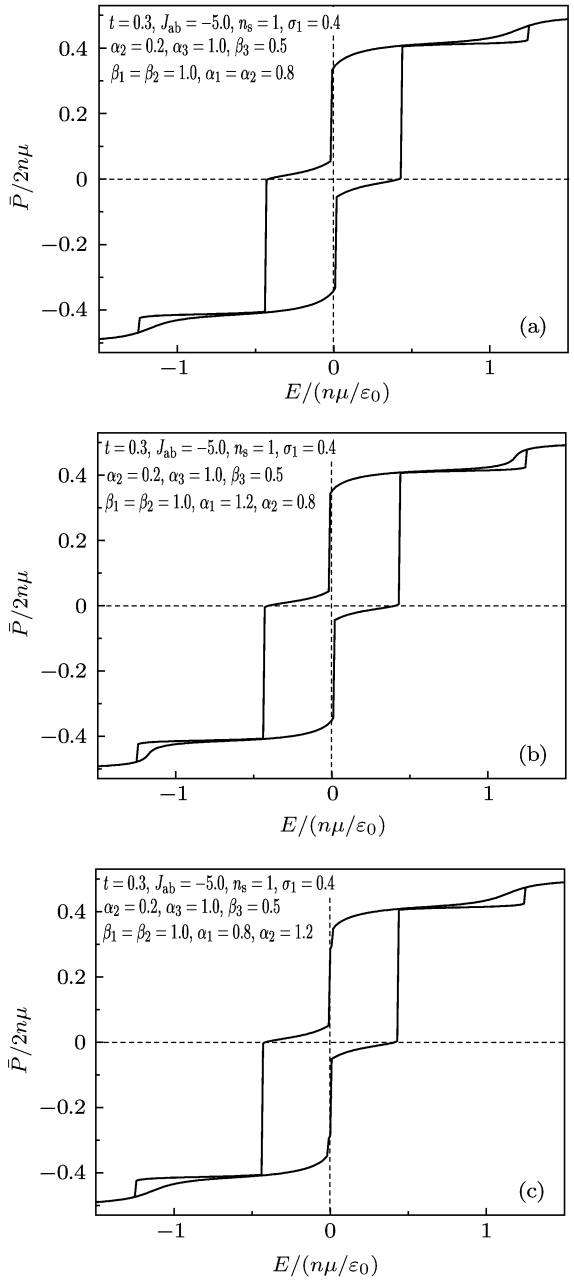


Fig. 4. The hysteresis loops of a bilayer film with various values of α_1 and α_2.

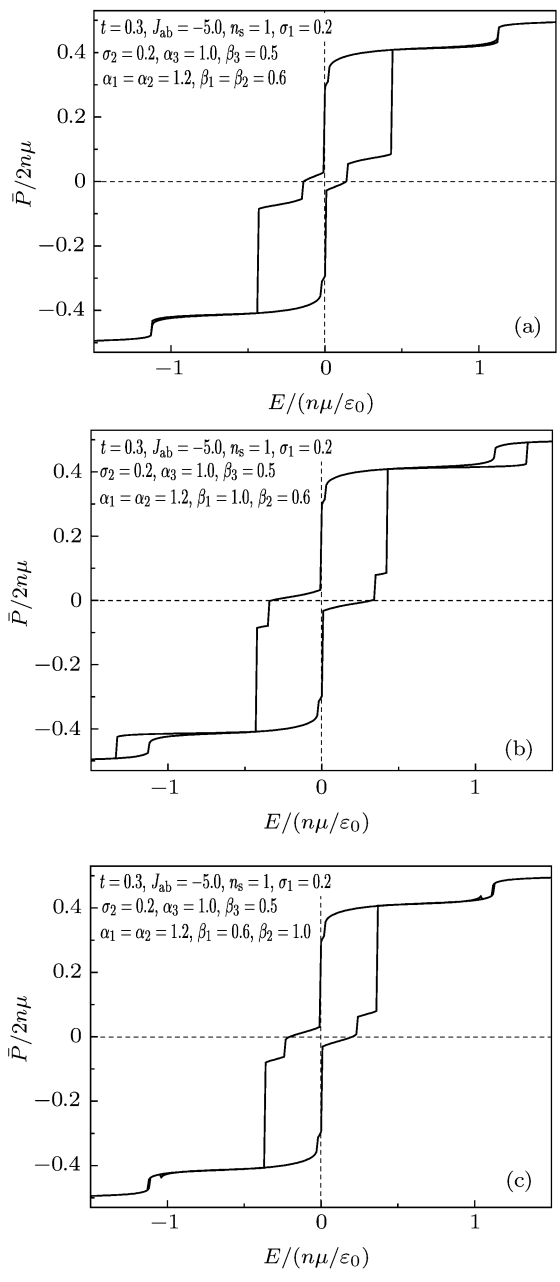


Fig. 5. Dependence of hysteresis loops on the different values of β_1 and β_2.

The hysteresis loops of a bilayer film with different α_3 and β_3 are shown in Fig. 6. It is known that the pseudo-spins system becomes disordered as the transverse field increases. In our selected system, the slab A is easier to be disordered than slab B. Thus, when the value of β_3 increases, namely, the transverse field in the surface transition layer in slab B augments, the hysteresis loops change unconspicuously, as shown in Figs. 6(a) and 6(b). However, as α_3 augments, that is to say, the transverse field in the surface transition layer in slab A increases, the two sides symmetric loops vanish but two steps at two side emerge. The two steps result from the flip of the interior layer and the lower surface transition layer (see Fig. 1) in slab B. The reason is similar to that of Fig. 3(d). As α_3 further increases, there will not be two steps at two sides (see Fig. 6(d)). Therefore, there are also two critical values: α_{3c_1} and α_{3c_2}. When $\alpha_{3c_1} < \alpha_3 < \alpha_{3c_2}$, the two steps at two sides will appear.

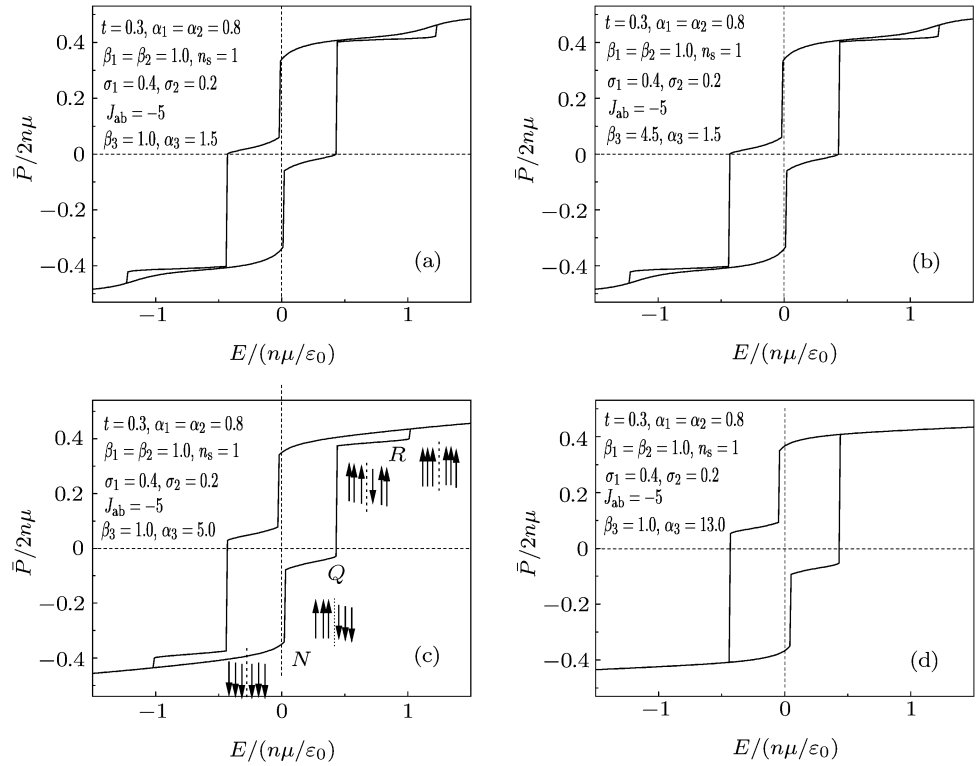


Fig. 6. The hysteresis loop s of a bilayer film with different values of α_3 and β_3.

Figure 7 describes the hysteresis loops of a bilayer film with different thicknesses of surface transition layer.

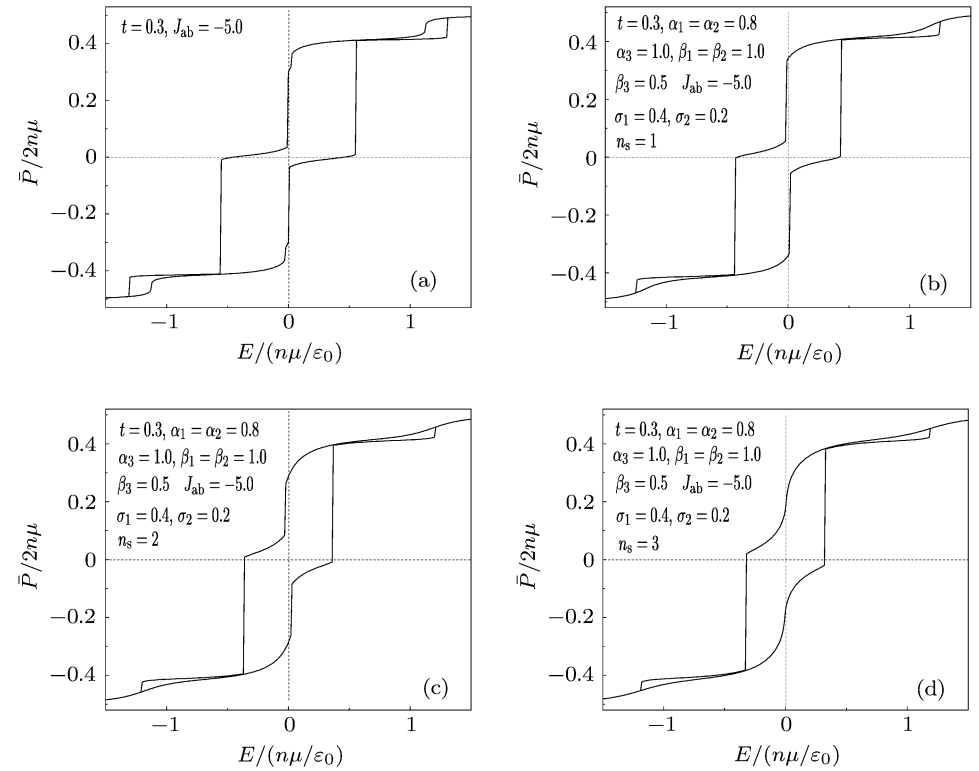


Fig. 7. The hysteresis loops of a bilayer film for different surface transition layers.

Figure 7(a) represents the case without the surface transition layers. Compared to Fig. 7(a), due to the existence of the surface transition layers, the size of the outer loops is small and that of the central loop becomes narrow horizontally. And the hysteresis loop of a bilayer film presents an inferior rectangular degree, which is disadvantageous to the applications of memory devices, in particular for the thick surface transition layer (eg. Fig. 7(d)). As the thickness of the surface transition layer increases, the area of the outer loops shrinks and that of the central loop reduces. The augment of the surface transition layer thickness, leading to the strong surface effect, makes the electric field easily change the polarisation of a slab.

The dependence of the hysteresis loops on the temperature is shown in Fig. 8. From Fig. 8, we can see that the size of the outer loops and that of central loop reduce with increasing temperature. This behaviour is in good agreement with that observed by Ranjith.[13] If the temperature is high enough, the outer loop will disappear and the central loop will become much narrow (see Fig. 8(c)). When $t = 1$, in other words, the temperature is the phase transition temperature, the P–E curve of a bilayer film becomes a curve of paraelectric polarisation as a function of the applied electric field. Here, both slab A and slab B are disordered.

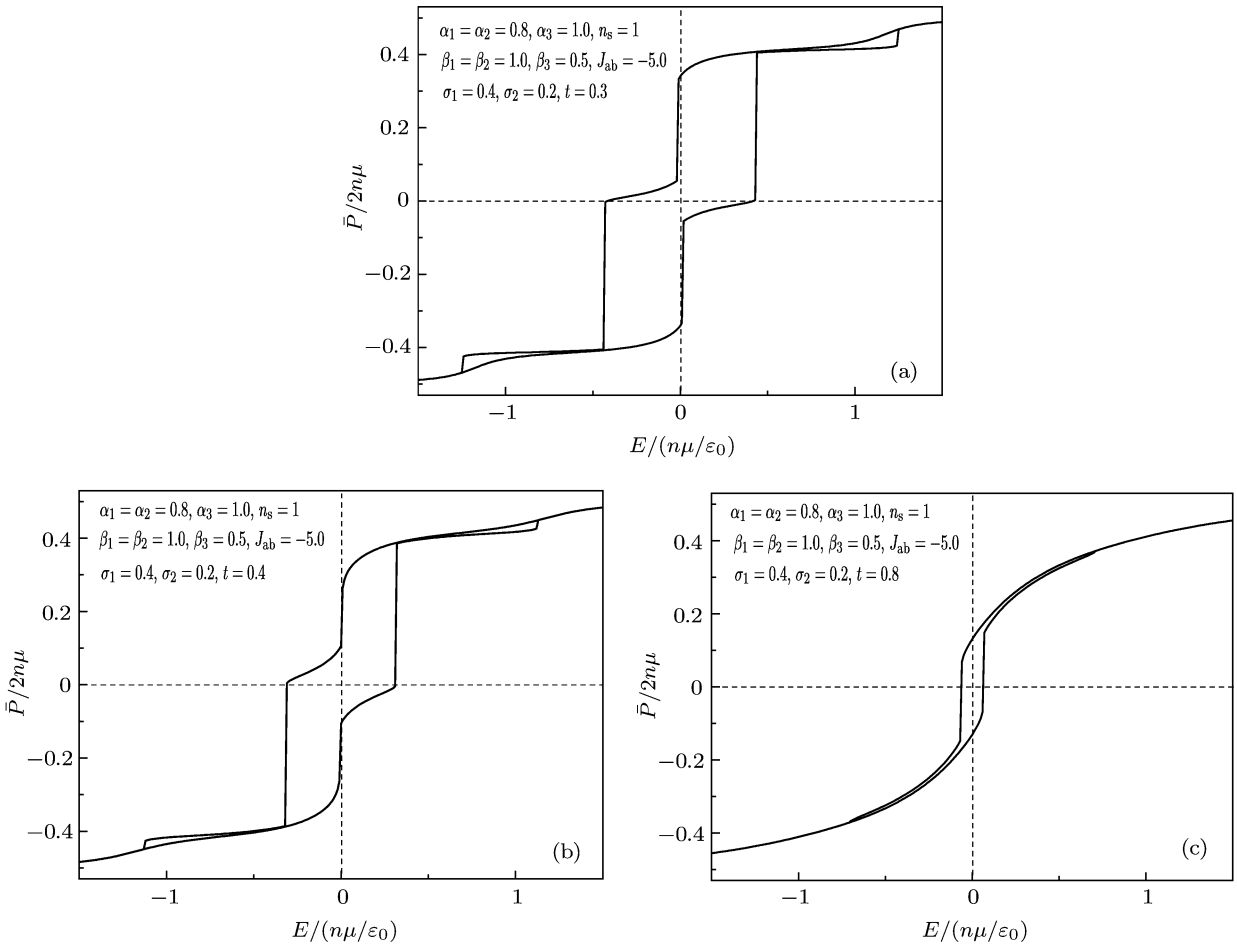


Fig. 8. The hysteresis loops of a bilayer film at different temperatures.

4. Conclusions

In summary, taking into account surface transition layers, we study the hysteresis loops of a ferroelectric bilayer film with an antiferroelectric interfacial coupling by use of the Ising model under the mean field approximation. Our results can be summarized as follows: (i) the effects of the surface transition layers on the hysteresis loops of a ferroelectric bilayer film should be considered; (ii) the existence of the surface transition layer makes the system have more ferroelectriclike characters, although the effect of the antiferroelectric coupling favours

antiferroelectriclike properties of the system; (iii) for a given thickness of the surface transition layer, there are two critical values of interactions for the stronger interactions of the bulk material and of transverse field for the weaker interactions of the bulk material, respectively. Between the two critical values, four steps appear in the central loop, the other cases will be two steps for two different constituent slabs.

Our results may provide some guidance in experiments, and correctly realize a multiple-state memory in practice for future.

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