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Size Effects of the Critical Temperature in Ferroelectric Thin Films*

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Abstract *The size effects of the critical behaviors for the systems of interacting spins are discussed extensively in literature. In this paper, the finite-size dependence of the critical temperature and susceptibility of the ferroelectric thin film are investigated numerically based on the four-state Potts model with the nearest-neighbor interactions between the dipole moments. The four orientations of the domains exist in the ferroelectric film and the movement of the domain walls determines the polarization switching process besides the boundary conditions of the film. The critical exponents are obtained and our investigations show that the boundary conditions play the important roles for the ferroelectric properties of the thin films and the critical behavior of the thin films strongly depends on the feature of the surface.*

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1 Introduction

One of the important problems in the study of thin ferroelectric films is the size effects of critical behavior. Although it has been investigated since the 1950's,[1,2] it is revisited due to the rapid developments of ferroelectric films and the applications, such as micro-electrical mechanical system, nonvolatile memory cell and optical wave-guide devices.[3,4] With the aid of modern deposition techniques, a quality layer-to-layer growth can be obtained, which permits the description of films with nearly flat surfaces by two-dimensional (2D) models. These 2D models have been adapted extensively for the theoretical studies. It becomes a powerful tool for the theoretical investigations of the finite-size effect in ferroelectric crystals and thin films where there are a lot of experimental results. Recently, Stachiotti showed from atomic-level simulations that the critical thickness for ferroelectricity in a free-standing $\mathrm{BaTiO_3}$ is 3.6 nm.[5] Junquera and Ghosez reported that the critical thickness for $\mathrm{BaTiO_3}$ film between two metallic $\mathrm{SrRuO_3}$ electrodes is 2.4 nm from first-principles calculations.[6] Lai *et al.* provided the detailed atomistic insight of the domain evolution of epitaxial ultrathin films under an applied electric field.[7]

We know that ferroelectricity is analogous to ferromagnetism in many ways. The second-order phase transition has been observed in monolayer magnetic films and many ferromagnetic materials. Even in the simplest 2D Ising model, the phase transition is possible in the 2D lattice as shown by Onsager.[8] Now it is clear that we can deal with the ferroelectric phase transitions in 2D lattice.[9] Ultra-thin crystalline films offer the possibility of exploring phase transitions in the crossover region between two and three dimensions. The second-order transition at lower temperature was observed in the thin film and this system should be considered as two-dimensional ferroelectrics[9] due to the near-absence of finite-size effects on the bulk transitions.

The earliest research on the ferroelectric phase transitions was from IBM group.[10−12] They considered the effect of electrodes and concluded that the critical temperature of the film was reduced by the depolarization from a transverse electric field. The size and surface effects on ferroelectric phase transitions have been extensively studied by using the Landau theory.[13,14] Lubensky and Rubin[15] discussed the phase transitions on mean-field theory where the effect of boundary polarization at the surface was considered. Tilley and Zeks[16] studied the Landau–Devonshire model, in which the bulk transition is of second order. Scott *et al.*[13] investigated the first-order phase transition based on the Landau phenomenological theory. Qu *et al.*[17] studied the phase transition of spontaneous polarization and the dielectric susceptibility of ferroelectric thin film based on Ising model in a transverse field. Aarao Reis[18] dealt with the critical behavior of the thin Ising films with noninteger mean thicknesses. Ong *et al.*[19] gave a detailed discussion of critical behavior of thin ferroelectric films where the polarization was in terms of Jacobi elliptic function. Recently, Baratkovsky and Levanyuk[20] studied ferroelastic phase transitions in thin epitaxial films and they found the formation of sinusoidal strain wave. Very recently, a thermodynamic model has been developed to describe the critical parameters in

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the ferroelectric transformation in the films based on the time-dependent Ginzburg–Landau equation in Ref. [21] where the difference between the superheating and supercooling transition temperature for the first-order transitions is found to be insensitive to the film thickness and surface boundary conditions.

An alternative approach of the phase transitions is based on the q-state Potts model.[22] This model exhibits temperature-driven first or second phase transitions, which has been extensively studied by both analytical and numerical approaches.[22−25] In order to study the influence of quenched random variables on phase transitions, the random bond and the random field Potts models[26] have been discussed recently using the double peak distributions of couplings and magnetic field. Chen *et al.*[27] undertook an extensive Monte Carlo investigation of the random-bond eight-state Potts model where a second-order phase transition exists for the pure case of the model. They found that this transition is of the same universality as the two-dimensional pure Ising model. From the finite-size scaling technique and conformal invariance Cardy and Jacobsen[28] showed a continuous transition with the random magnetic exponent which varies with q. But the correlation length exponent related to the critical temperature is consistent with the pure Ising model within computational error. Then, using the cluster flipping algorithm, Chatelain and Berche[25] concluded that the critical exponents, corresponding to susceptibility and random magnetization, were different from those in the pure Ising model through large-scale Monte Carlo simulations. Paredes V and Valbuena[29] focused on random five-state Potts model exhibiting a weak first-order phase transition, and they got that the presence of disorder also decreases the transition temperature. Olson and Young[30] verified that there was only a single exponent in describing the divergence of the correlation length in the critical behavior of the random q-state Potts model.

The presence of domain structures is a common feature of ferroelectric films below the critical temperature. The knowledge on the domain dynamics is crucially important for controlling the properties such as switching, permittivity, and piezoelectricity, etc. The orientation of dipoles changes in ferroelectric transitions. In principle, the corresponding microstructure of the ferroelectric phase contains various of domains separated by domain walls in the absence of any external field. Experimentally it was found that 90° domains are predominant in most tetragonal ferroelectrics.[31] This domain formation has been simulated by using Landau theory.[32] Energetics and geometry of 90° domains structures were investigated in the epitaxial ferroelectric thin films in Ref. [33]. Emelyanov and Pertsev studied the abrupt changes and hysteretic behavior of 90° domains with misfit dislocations in Ref. [34].

As electronic devices become smaller and smaller, it is highly required to understand phenomena occurring in nanoscale, which are much different from bulk properties. In ferroelectric ultrathin film systems, ferroelectric phase transition plays an important role in ferroelectric domain dynamics. It is very important to study the critical behavior of the ultrathin film based on the q-state Potts model by considering the domain structures. The critical behavior of the ferroelectric thin films strongly depends on the feature of the surface. The understanding of the ferroelectricity in ultrathin ferroelectric systems would help us in the development of ultrathin ferroelectric devices such as ferroelectric ultrathin film capacitors and ferroelectric memories.

In this paper, we are going to study the critical behavior induced by the paraelectric-ferroelectric phase transition, which will be referred as the phase transition in subsequent section. Our investigation is based on the four-state Potts model with the nearest-neighbor interactions between the dipole moments. The thickness dependence of the critical temperature is discussed and the critical exponents are obtained for the thin film. Our result shows that the critical behavior of the ferroelectric thin film is different from that of the bulk. The domain structures of the ferroelectric thin films, such as the $\mathrm{Pb(Zr}_x\mathrm{Ti}_{1-x})\mathrm{O}_3$ (PZT) film, have the narrow stripes of 90° domains and the wedged-shape 90° domains. The statics and dynamics of 90° domains in ferroelectric thin films can be affected considerably by the interaction with misfit dislocations. As a possible microscopic mechanism of the Barkhausen effect occurring in ferroelectric thin films during the polarization reversal, the steplike movements of 90° domain walls over the misfit dislocations are proposed in Ref. [34]. The characteristic etch patterns in PZT with boundary parallel to [100] are also 90° domains. In the next section, we introducing the model describing the ferroelectric thin films switching through the 90^o domains. The temperature dependence of susceptibility of the model is investigated in Sec. 3. We get that the correlation length exponent is close to pure Potts model with periodic boundary condition. But the critical exponent associated to the susceptibility is different from the pure case of the model. Our investigation shows that the critical behavior of the thin film strongly depends on the properties of the surface. Finally, we give the conclusions and remarks in Sec. 4.

2 The Model

There have been a number of efforts to study the energetics of domain structures in ferroelectric thin films and these domain structures are usually formed below their Curie temperature T_c. A two-dimensional ferroelectric thin film based on four-state Potts model with the nearest-neighbor interactions can be considered as a two-dimensional array of dipoles $(N_x \times N_z)$ in x-z plane, where

z is the thickness direction. There are four mutually perpendicular orientations in our model. These orientations of the dipoles are denoted by the states such as $A(\uparrow)$, $C(\downarrow)$, $B(\rightarrow)$, $D(\leftarrow)$ for up, down, right and left dipoles, respectively. These are the only possible directions and hence domains, in a single crystalline thin film or in polycrystalline film with a dominant crystal orientation. The dipole moment for a cell located at the position (i, j) inside the film is given by $P_{i,j} = P_0 S_{i,j}$ where $S_{i,j}$ is called a pseudo-spin. The initial state of these spins in each cell is randomly generated to one of these four states. The system Hamiltonian is

$$H = -J \sum_{i,j} \sum_{i',j'} \bar{\boldsymbol{S}}_{i,j} \cdot \bar{\boldsymbol{S}}_{i',j'} - \sum_{i,j} \bar{\boldsymbol{E}}_j \cdot \bar{\boldsymbol{S}}_{i,j} \,, \tag{1}$$

where J is the coupling coefficient of the interaction between the nearest-neighbor pseudo-spins $\boldsymbol{S}_{i,j}$ and $\boldsymbol{S}_{i',j'}$, $\bar{\boldsymbol{E}}_j$ is the effective electric field $\bar{\boldsymbol{E}}_j = \bar{\boldsymbol{E}}'_j P_0$ related to the external electric field $\boldsymbol{E}'_j$. Only the nearest neighbors are included in the summation in the model. When these spins are taken as unit vectors, their product gives the following results: (a) +1, when they are parallel; (b) −1, when they are antiparallel; (c) 0, perpendicular. The electric field is along z-direction. Thus only the spins in states A and C contribute to the second term in Eq. (1). The state A gives the value $-E_j$ while the state C gives the value $+E_j$.

Now the critical exponents of the ferroelectric thin films can be obtained by using the Monte Carlo simulation. As in the conventional Ising Model, a spin $S_{i,j}$ is randomly chosen at each step. For the laminar 90° domain structures, there exists a linear relation in the range of large film thicknesses in the epitaxial films and ferroelectric ceramics, which normally obey the square root law of the thickness.[33,35,36] To animate the switching through the domain wall movement, whether a spin is allowed to rotate is tested by the following two conditions simultaneously.

(i) The selected spin is in the domain boundary or in the electrode/film boundary. This condition can be expressed by the following expression:

$$S_{i-1,j}S_{i,j} + S_{i+1,j}S_{i,j} + S_{i,j-1}S_{i,j} + S_{i,j+1}S_{i,j} \neq 4 \,. \tag{2}$$

(ii) When the above condition is satisfied, the change in Hamiltonian $\Delta H = H'(S') - H(S)$ is evaluated after the related spin has been rotated 90°, $S_{i,j} \rightarrow S'_{i,j}$. As in the conventional Ising model, the rotation of the spin is allowed if $\Delta H < 0$ or $r < \exp(-\Delta H/kT)$, where r is a random number in the interval of [0,1]. Unlike the conventional Ising model, the rotating of the related spin is 90°, with equal probabilities in two directions: clockwise or anti-clockwise. The polarization P_i can thus be evaluated by taking the average value of the dipole moments over all cells. The dielectric susceptibility χ of the thin film is given by[37]

$$\chi = \frac{ld}{kT} (\langle P^2 \rangle_T - \langle |P| \rangle_T^2) \,, \tag{3}$$

where P is the polarization, l is the length of the film, and k is the Boltzmann constant. The order parameter $\langle |P| \rangle_T$ is given by $\langle |P| \rangle_T = (\sum_{i=1}^{N} |P_i|)/N$ with simulating the polarization N times and $\langle P^2 \rangle_T = (\sum_{i=1}^{N} P_i^2)/N$ where a small field is added to the film at the temperature T. Our simulation is based on the fact that this dielectric susceptibility shows a critical divergence in thermodynamic limit and it only reaches a maximum of finite height in the finite system,[37] that is,

$$\chi_{\max} \sim d^{\gamma_c} \,, \tag{4}$$

when $T = T_c(d)$. The corresponding critical temperature $T_c(d)$ for maximum susceptibility shifts with the thickness d for a fixed film length. The correlation length exponent is given by

$$T_c(\infty) - T_c(d) \sim d^{-1/\nu} \,, \tag{5}$$

where ν is also called thermal exponent.[18,28] The detailed results are shown in the following section.

3 Critical Exponents of Thin Film

The critical exponent ν for Curie temperature is obtained from Eq. (5). Two kinds of boundary conditions have been used in the simulations. For the first kind the free boundary condition applies on the top and bottom surface and periodic boundary condition applies on two edges of the film. We denote this case as Boundary Condition A (BCA). For the second case, periodic boundary condition applies on all surfaces of the film, denoted as Boundary Condition B (BCB). For the asymptotic behavior of the critical temperature with the film thickness, it is independent of the contact conditions, such as the dielectric contacts and short-circuit electrodes.[21] As usual situation in the simulation, we work in units so that the related parameters, such as temperature and field, are dimensionless and we use the small field in the simulated calculations. The film length and thickness are expressed by the corresponding numbers of layers. When we take the values of the lattice constants, for example, of PZT, as 0.41 nm approximately,[38] the film length and thickness are presented dimensionally. For instance, the film length l being 450 unit cells in Fig. 1 means that the length is 184.5 nm for the PZT film. And the different lattice constants of the different ferroelectric materials may give the different values of the length and thickness for the same number of the layers.

Figure 1 depicts the dependence of the critical temperature on the film thickness with a fixed film length $l = 450$ (number of layers) representing 450 unit cells, where the small field 0.3 exists along the z-direction. The simulated result shows that the correlation length exponent $\nu = 1.00 \pm 0.05$ under BCA. It is very interesting that this fitted critical exponent agrees with random bond Potts models[30] within the computational errors. Under the case BCB, the fitted critical exponent becomes $\nu = 0.67 \pm 0.03$ (See Fig. 2), approaching the value for pure four-state Potts model.[22] This result implies that the correlation length exponent is very sensitive with the boundary conditions and the exponent of thin film is different from the value of a bulk sample. We know that the thermal exponent is 1 for the thin film and 0.5 for the thick film when the lattice relaxation weakens the polarization on the surface.[21] Then, our simulations mean that the BCA is a good choice for describing the ferroelectric behavior of the thin films and BCB is more suitable for the thick films.

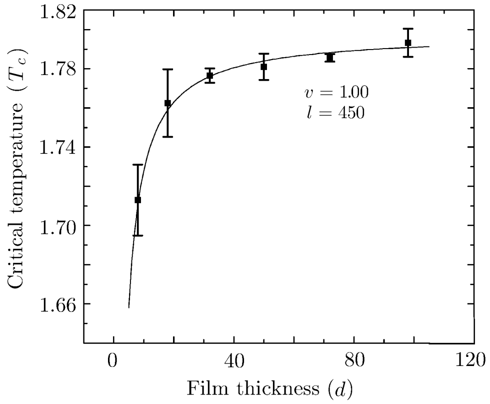

Fig. 1 Thickness dependence of the Curie temperature with the film length $l = 450$ (number of layers). It gives that the exponent $v = 1.00 \pm 0.05$ under the case BCA.

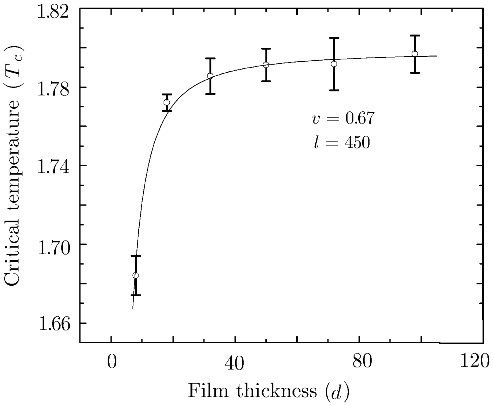

Fig. 2 Thickness dependence of the Curie temperature for the case BCB. It gives $v = 0.67 \pm 0.03$.

Figure 3 shows the polarization profile throughout the thin film with the length $l = 162$. Their critical temperatures T_c are 1.712, 1.753, and 1.778 corresponding to the thickness 8, 13, and 18 (number of layers), respectively. The boundary condition is the same as the one in Fig. 1. In such films, Curie temperature shifts to lower temperatures compared with the bulk value. This is consistent with the result given by Tilley and Zeks[39] by using the Landau theory and also by Wang *et al.*[40] using a transverse Ising model. This figure also reveals that the surface polarization is smaller than the value deep inside the film.

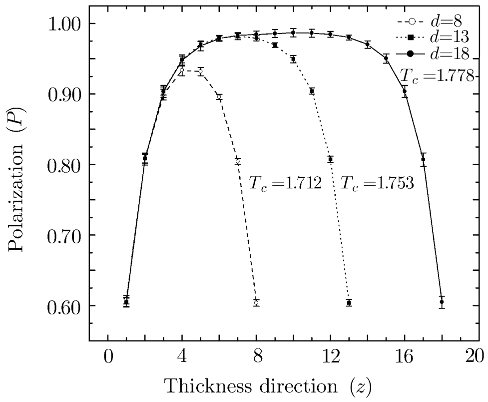

Fig. 3 The distributions of the polarization in the thin film with the fixed length $l = 162$ (number of layers). The corresponding critical temperatures are 1.712, 1.753, and 1.778 when the film thickness being 8, 13, and 18 (number of layers), respectively.

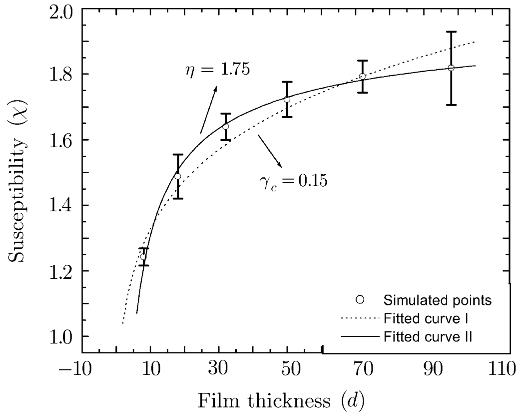

Fig. 4 Thickness dependence of the dielectric susceptibility under the case BCB. Two kinds of the power-law fits are corresponding to the form (4) for fitted curve I and the form (6) for curve II. They give that $\gamma_c = 0.15 \pm 0.01$ (dotted line) and $\eta = 1.75 \pm 0.04$ (solid line).

The critical exponent related to the susceptibility has also been obtained. In Fig. 4, the finite size effect of the susceptibility at the critical point is presented under BCB. Fitting the curve using Eq. (4) gives that $\gamma_c = 0.15 \pm 0.01$ (dotted line). It is much smaller than the value derived from a bulk sample but larger than that of the local surface susceptibility, $\gamma_{11}/\nu = 0.099 \pm 0.009$.[25] It is revealed that γ_c of the thin film varies between the bulk and surface case. The exponent is approaching to the surface situation when reducing the thickness of the ferroelectric film.

If the susceptibility at Curie temperature is fitted by an alternative relation:

$$\chi_d = a - bd^{-1/\eta}\,, \tag{6}$$

where a, b are fitted parameters, and η is the critical exponent similar to the form as in Eq. (5). The value $\eta = 1.75 \pm 0.04$ was thus obtained. The fitted curve is shown in Fig. 4 for the case BCB. On the other hand, $\gamma_c = 0.16 \pm 0.01$ was obtained for the case BCA, as shown in Fig. 5, and $\eta = 1.75 \pm 0.03$. Figure 6 also reveals that the divergence of the susceptibility for the thin film is increased under free boundary condition. These results of the exponents γ_c and η are presented in Table 1.

Table 1 The critical exponents associated with the dielectric susceptibility of the film with the different boundary conditions.

Boundary conditions	Eq. (4)	Eq. (6)	Figures
BCB	$\gamma_C = 0.15 \pm 0.01$ Curve I	$\eta = 1.75 \pm 0.04$ Curve II	Fig. 4
BCA	$\gamma_C = 0.16 \pm 0.01$ Curve III	$\eta = 1.75 \pm 0.03$ Curve IV	Fig. 5

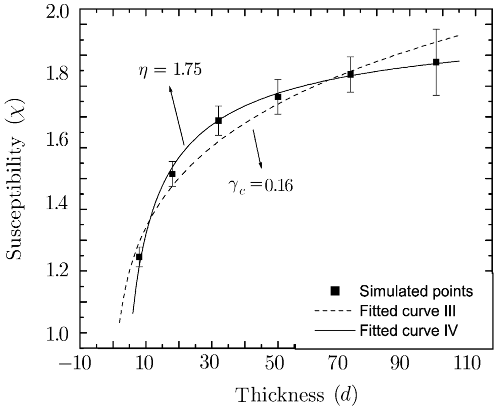

Fig. 5 Thickness dependence of the susceptibility corresponding to the case BCA. The fitted results give that $\gamma_c = 0.16 \pm 0.01$ (curve III) and $\eta = 1.75 \pm 0.03$ (curve IV).

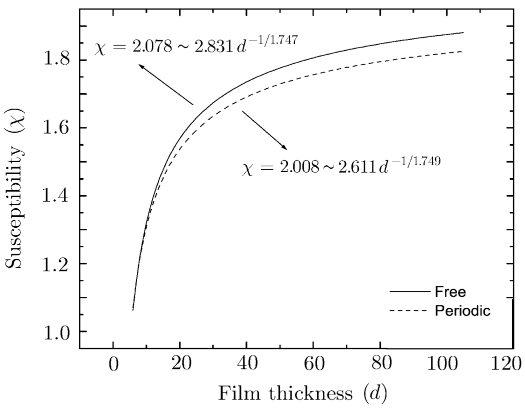

Fig. 6 The comparison of the thickness dependence of the susceptibility with different boundary conditions in the top and bottom surface of the film.

4 Remark and Conclusion

In this paper, we have simulated the critical behavior of the ferroelectric thin film by using the four-state Potts model with the nearest-neighbor interactions between the dipole moments. In fact, the domain patterns in the film provide the means to release stored electric and elastic energy at the energetic expense of domain walls formed between variants and the local stresses in the substrate. Mechanical sources of the long-range internal stresses in the strained epitaxy with a 90° domain structure are localized on the film/substrate interface.[33] Some domain patterns will be prevented from the formation due to the strain in the thin film. The polarization gradients, due to the coupling of the stress field of the dislocation, result in the depolarizing fields which suppress the polarization and introduce the dead layers that severely degrade ferroelectric properties.[41] This detrimental effect of the dead regions will be enhanced in ferroelectric thin films and should play the extrinsic role in size effect of the ferroelectrics. It needs to be discussed further based on the details of the depolarization field and the detrimental layers.

In summary, by simulating the critical behavior of the ferroelectric thin film with the use of the four-state Potts model, we found that the boundary condition plays an important role in the ferroelectric features of the films. Free boundary condition increases the divergence of the susceptibility and makes the correlation length exponent approaching to the random bond Potts model. On the other hand, the periodic boundary condition preserves the exponent ν consistent with the pure model for the thin film. It is a good choice of the free boundary condition for the ferroelectric properties of the thin films. When the thickness becomes bigger, the periodic boundary condition is more suitable to simulate the ferroelectric behavior of the films. The exponent associated with dielectric susceptibility varies between the surface value and the bulk value. This means that the critical behavior of the thin film strongly depends on the properties of the surface. The reduction of surface polarization is correlated with the reduction of Curie temperature when the thickness d decreases. When an alternative functional dependence of susceptibility similar to that for critical temperature is adapted, the critical exponent 1.75 ± 0.04 was obtained for the thin film.

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