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EFFECTS OF LONG-RANGE INTERACTIONS ON THE FERROELECTRIC FILM*

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The effect of long-range interactions on the spontaneous polarization and the Curie temperature of the ferroelectric film is investigated by use of the Landau theory. On the assumption that the nearest-neighbour interaction remains constant, we find that the spontaneous polarization and the phase transition temperature increase with the enhancement of the long-range interactions. In the case of positive extrapolation length, the critical thickness of the ferroelectric film, in which a size-driven phase transition occurs, decreases with the enhancement of the long-range interactions.

Keywords: long-range interaction, ferroelectric film
PACC: 7780

I. INTRODUCTION

Thin ferroelectric films have attracted much attention due to the potential applications to various electronic devices, such as dynamic random access memory.[1] Many researchers have focused their attention on the functionally graded ferroelectric thin films[2] or ferroelectric multi-layers, in which different layers have different polarizations, and the interactions of the polarization between different layers play important roles in the properties of the films. Much theoretical work has been done based on the Landau theory of phase transition. Kretschmer and Binder[3] introduced Landau theory into the study of surface effects of the ferroelectric thin film. Scott, Zhong and co-workers[4−8] discussed the change of polarization and Curie temperature under the same theoretical frame.

In order to take account of the polarization inhomogeneity and the surface effect, a gradient term in the polarization and a parameter δ, called the extrapolation length, were introduced into the free-energy expression of the film. It was shown that the polarization and the Curie temperature of the ferroelectric film decreases (increases) with the decrease of the thickness of the ferroelectric film for positive (negative) values of δ, and in the case of positive extrapolation length, a size-driven phase transition occurs. In order to solve the Euler–Lagrange equation of the polarization with a given boundary condition, the finite-difference scheme was used. Under the assumption that the polarization has a distribution along the z-axis (perpendicular to the film plane), the film was divided into several layers along the z-axis, having different polarizations. However, the interactions between these layers of inhomogeneous polarization were not taken into account. As is well known, in ferroelectrics, long-range interactions are thought to be dominant, a fact which especially should be considered in the study of finite-size ferroelectrics.[9−11] In this paper, we introduce the long-range interactions between the layers of inhomogeneous polarization in the free-energy expression of the ferroelectric film to investigate their effects on the spontaneous polarization and the Curie temperature of the film. The transverse Ising model (TIM) was first applied by Gennes *et al.*[12] to describe the order–disorder-type ferroelectric phase transition. The TIM with long-range interactions has also been used to describe the ferroelectric film.[9] However, the materials of most interesting functional ferroelectric films are typically displacive. This paper provides an alternative way to investigate the effects of long-range interactions of the polarization on the phase transition of ferroelectric films.

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Ⅱ. THEORY

Supposing the inhomogeneity of the ferroelectric film is along the z-direction and the polarization in the film plane is uniform, for a ferroelectric of the second-order phase transition, the Landau-like free-energy expression for the film with long-range interactions of the polarization can be written as[10]

$$\frac{F}{S}=\int_{-L/2}^{L/2}\left[\frac{1}{2}A(T-T_{C\infty})P^2(z)+\frac{1}{4}BP^4(z)+\frac{1}{2}D\left(\frac{dP}{dz}\right)^2-\gamma P(z)\right]dz+\frac{1}{2}D\delta^{-1}(P_+^2+P_-^2), \quad (1)$$

where δ is the extrapolation length which reflects the difference between the surface and the interior of the film, $T_{C\infty}$ is the bulk Curie temperature, P_+ and P_- are the polarization at $z=\frac{1}{2}L$ and $z=-\frac{1}{2}L$, respectively. $\gamma=\int_{-L/2}^{L/2}\frac{JP(z')}{r_{ij}^{\eta}}dz'$, where $r_{ij}=|i-j|=1,2,3,...$ is the distance (in crystallographic units) between the planes i and j in the film. The term $\gamma P(z)$ corresponds to the contributions of long-range interactions of polarization between different planes of the film. η is introduced to describe the magnitude of the long-range interaction. The parameter J represents the coefficient of the short-range interaction, because for the nearest-neighbour interaction at any η, $r_{ij}^{n}=1$, thus $J_{ij}=J$. In this paper, we focus our attention on the effects of the long-range interaction of the polarization. So, the value of the coefficient J is fixed and only the parameter η is changed to investigate the effects of the long-range interaction on the phase transition of the film. From Eq.(1), we can see that the smaller the parameter η is, the stronger the long-range interaction. L and S are the thickness and the area of the film, respectively. A, B, C and J are positive coefficients. Here, the free energy of the paraferroelectric phase is taken to be zero.

For the ferroelectric film of the first-order phase transition, the free-energy expression Eq.(1) can be modified as[6,10]

$$\frac{F}{S}=\int_{-L/2}^{L/2}\left[\frac{1}{2}A(T-T_{0\infty})P^2(z)+\frac{1}{4}BP^4(z)+\frac{1}{6}CP^6(Z)+\frac{1}{2}D\left(\frac{dP}{dz}\right)^2-\gamma P(z)\right]dz+\frac{1}{2}D\delta^{-1}(P_+^2+P_-^2), \quad (2)$$

where the coefficient B is negative and $T_{0\infty}$ is the bulk Curie–Weiss temperature. Minimizing the free energy F with respect to the polarization, we obtain the following Euler–Lagrange equations

$$D\frac{d^2P}{dz^2}-A(T-T_{C\infty})P-BP^3+\gamma=0 \quad (3)$$

and

$$D\frac{d^2P}{dz^2}-A(T-T_{0\infty})P-BP^3-CP^6+\gamma=0, \quad (4)$$

for the ferroelectric films of the second-order and first-order phase transitions, respectively. The boundary condition is

$$\frac{dP}{dz}=\mp\frac{P}{\delta}, Z=\pm\frac{1}{2}L. \quad (5)$$

Equations (3) and (4) can be solved by using the finite-difference method. At every thickness, the polarization distribution for different temperatures was obtained. Then substituting the polarization into the free energy expression, we can obtain the free energy of the ferroelectric film, and by equating the free energy to zero, we get the Curie temperature of the film.

Ⅲ. RESULTS AND DISCUSSION

The spatial distribution of the polarization at selected parameters (η) of long-range interactions at zero temperature is calculated first for the ferroelectric film with a second-order phase transition (see Figs.1 and 2). Figs.1(a) and 1(b) are the polarization distributions along the z-axis for positive and negative δ, respectively, at a fixed thickness of the film. The phenomenological parameters are taken to be: $A=1$, $B=1$, $D=100$, $\delta=50$, $J=2.5$, $L=10$.

From Fig.1, we can see that for a positive value of δ, the surface polarization of the film is smaller than the internal polarization, with the opposite being true for negative δ. Whether the value of δ is positive or negative, the polarization always increases with the decrease of the parameter of long-range interaction (η). This is reasonable, as the ferroelectric film tends to be ordered with the enhancement of the long-range interaction, and the smaller the parameter η, the stronger the long-range interaction.

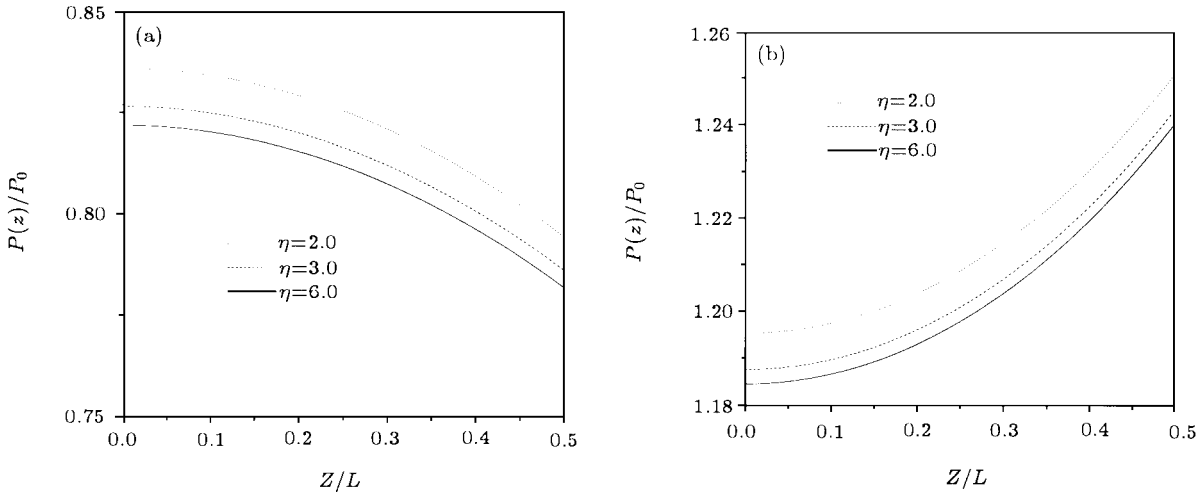


Fig.1. Depth profile of the polarization for a ferroelectric film with a second-order phase transition for selected parameters (η) of the long-range interactions when (a) $\delta = 50$ and (b) $\delta = -50$ at zero temperature. P_0 is the spontaneous polarization of the bulk ferroelectric at zero temperature.

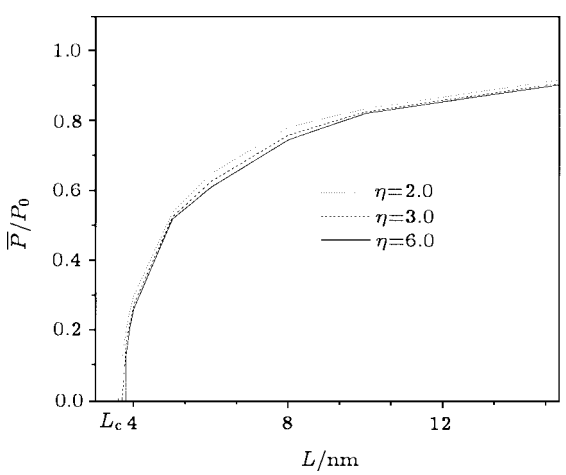


Fig.2. The average polarization as a function of the thickness of a ferroelectric film with a second-order phase transition for selected parameters (η) of the long-range interactions at zero temperature.

The average polarization of the ferroelectric film as a function of the thickness of the film for a positive value of the extrapolation length is plotted in Fig.2. A size-driven phase transition is found, and a critical thickness of the film exists below which the film presents a paraferroelectric phase at zero temperature, and the critical thickness (L_c) decreases when the long-range interactions strengthen.

For the ferroelectric film with the first-order phase transition, the parameters adopted in this paper are appropriate to the most extensively studied ferroelectrics with the first-order phase transition $BaTiO_3$, and the coefficients A, B, C, D, δ, $T_{0\infty}$ and $T_{C\infty}$ are taken from Ref.[12], namely: $A = 6.66 \times 10^5$, $B = -3.56 \times 10^9$, $C = 2.7 \times 10^{11}$, $D = 4.5 \times 10^9$, $\delta = 1 \times 10^{-9}$, $T_{0\infty} = 391$ and $T_{C\infty} = 403$ in SI units.

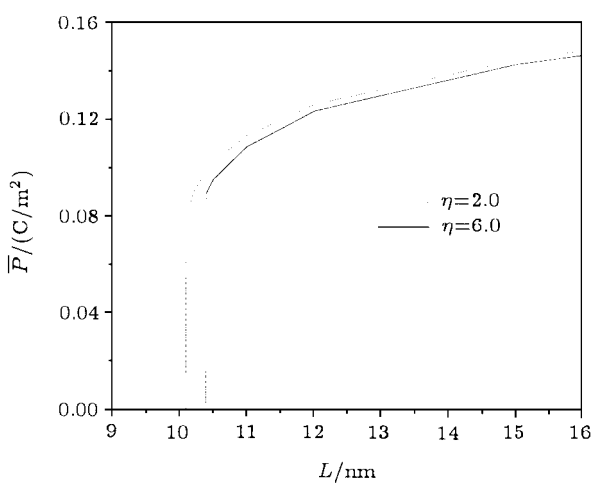


Fig.3. The average polarization as a function of the thickness of the ferroelectric film with the first-order phase transition for selected parameters (η) of the long-range interactions at zero temperature.

In Fig.3, we plot the average polarization of the film as a function of the thickness of the film at zero temperature. Different from the ferroelectric film of the second-order phase transition, the polarization first decreases with the decrease of the thickness and then disappears suddenly at a critical thickness, and the long-range interaction reduces the value of the critical thickness. As is known, the critical thickness without considering interactions of the polarization ($\gamma = 0$) is 11nm.[13]

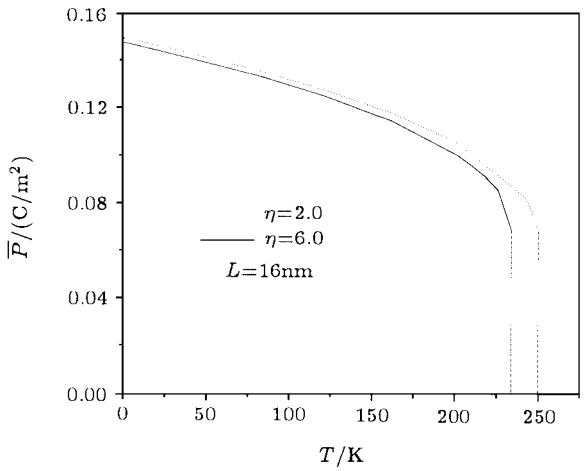


Fig.4. The average polarization of the ferroelectric film with the first-order phase transition as a function of the temperature at a fixed thickness L=16nm.

It is shown in Fig.4 that the average polarization decreases with the increase of the temperature for a fixed thickness of the film (L=16nm). Under the same temperature, the stronger the long-range interactions, the higher the value of polarization. It can be seen from Fig.4 that the long-range interaction raises the phase transition temperature. This can be seen clearly in Fig.5, where we give the curve of the phase transition temperature (T_C) versus the thickness of the film (L). The phase transition temperature increases with the increase of the thickness, and T_C approaches the bulk phase transition when the thickness is large enough. In summary, we have considered the influences of the long-range interaction of the polarization on the polarization, the phase transition temperature, and the critical thickness of the ferroelectric film. We conclude that the enhancement of the long-range interaction will increase the polarization and the phase transition temperature of the ferroelectric film, and decrease the critical thickness of the ferroelectric film.

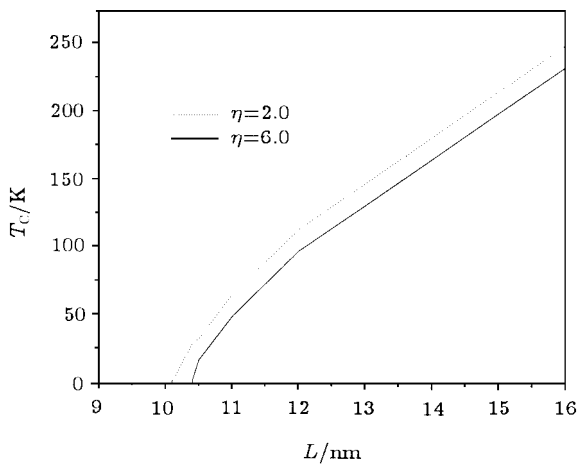


Fig.5. The Curie temperature of the ferroelectric film with the first-order phase transition as a function of the thickness for different long-range interactions (η).

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