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# Theoretical study on piezoresponse of ultrathin ferroelectric films

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Piezoelectric effect is crucial in some nano devices, but it usually decreases with the reduction of film thickness. A comprehensive study of the nonlinear piezoresponse under an applied voltage has been carried out within the framework of the Landau phenomenological theory. For epitaxial heterostructures, polarization exists even below the critical thickness of a few atomic layers due to the large compressive strain provided by the substrate. Piezoelectric coefficient could be very large in the vicinity of the critical thickness due to the appearance of a dielectric susceptibility peak at the phase transition point. Under an applied voltage, the susceptibility peak is reduced and broadened, accompanying a nonlinear piezoresponse even below the critical thickness. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4757946>]

## I. INTRODUCTION

Piezoelectricity plays an important role in many applications, including nano and quantum devices.<sup>1-8</sup> For instance, surface charges induced by the piezoelectric effect form an internal bias field, which affects the distributions of the charges and holes in semiconductors;<sup>9</sup> the electron tunneling behavior in ferroelectric tunnel junctions can be modulated by piezoelectric strain that changes the effective barrier thickness and electron mass.<sup>10</sup> However, the piezoelectric coefficient in nano scale is still ambiguous. Usually, it is thickness dependent, ranging from about 2–5 pm/V for less than 6 nm thick films detected by a conductive-tip AFM<sup>8</sup> to 60 pm/V for a 30 nm thick film.<sup>11</sup> In general, the piezoelectricity, similar to ferroelectricity, degrades with the decrease of film thickness due to various intrinsic and extrinsic effects.<sup>12</sup>

From a theoretical point of view, piezoelectricity in high-quality epitaxial ultrathin films is mainly dependent on the magnitude of the polarization, dielectric properties, defects, and microstructures.<sup>13</sup> With the counterbalance of applied strain, ferroelectricity could exist in films of only several atomic layers thick. In the vicinity of the critical thickness, there is a dielectric peak, indicating a thickness driven paraelectric to ferroelectric phase transition. Piezoelectric coefficient is expected to have a sharp increase near the critical thickness and disappears below it.<sup>14</sup>

In nano scale heterostructures, ferroelectric properties are significantly affected by (1) size effect due to the termination or distortion of long range order; (2) short range effect induced by band coupling and ionic displacement within several atomic layers from the interface of film and electrodes;<sup>15</sup> (3) depolarization induced by the incomplete compensation of bound charges at the interface due to the existence of electron distribution within a certain width from the interface inside the electrodes;<sup>16</sup> and (4) the effect of various fields, such as mismatch strain, temperature, and electric field.<sup>17,18</sup> These factors will also affect the dielectric and piezoelectric properties because of their relationship with the polarization.

In this work, we studied the piezoresponse of the film under an electric field with the consideration of imperfect conductor electrodes. Under an applied voltage, polarization and piezoelectricity can present in an ultrathin film even below the critical thickness with a broadened dielectric peak.

## II. THERMODYNAMIC MODEL

We focused on a single domain PbTiO<sub>3</sub> sandwiched between two symmetric SrRuO<sub>3</sub> electrodes on a thick substrate. The thickness of the film and each electrode were  $l_e$  and  $l$ , respectively. The origin of the axis was set at the interface of the left-side electrode and the film as shown in Fig. 1. The thick substrate can be considered rigid, thus, the in-plane dimensions of the thin film were totally constrained by the lattice mismatch strain induced by the substrate, and the degradation of the polarization induced by the size effect is significantly counterbalanced by the compressive strain, resulting in an enhancement of the polarization component perpendicular to the surface of the film. Since we are interested in ultrathin films with thickness less than 6 nm, the compressive strain from the substrate can be assumed to be uniform throughout the film.<sup>19</sup>

### A. Electrostatic and elastic energies

With the modification of first principle calculations, the Landau theory can be applied to films with thickness in

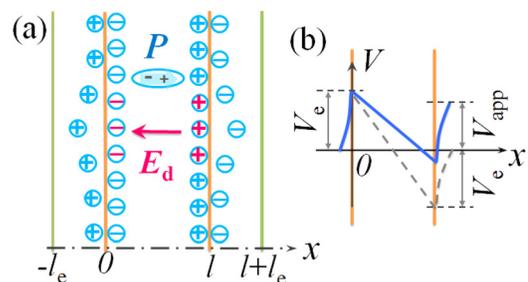


FIG. 1. (a) Illustration of incomplete charge distribution and formation of depolarization field. (b) Voltage distributions in the electrodes and film with and without applied voltage.

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nanometer regime.<sup>15,20</sup> Therefore, we adopted this method to study the polarization, dielectric, and piezoelectric properties in ultrathin films. With the application of an external electric field, the total polarization of a ferroelectric material includes spontaneous  $\mathbf{P}$  due to the ionic displacements results from the ferroelectric phase transition, and an induced  $\mathbf{P}_E$  by the applied electric field.<sup>21</sup> The electric displacement can be expressed as<sup>22</sup>

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}_E + \mathbf{P} = \varepsilon_0 \mathbf{E} + \chi_E \mathbf{E} + \mathbf{P} = \varepsilon_b \mathbf{E} + \mathbf{P}, \quad (1)$$

where  $\varepsilon_0$  is the vacuum dielectric permittivity,  $\chi_E$  is the linear field-dependent dielectric susceptibility of the reference dielectric material,  $\varepsilon_b = \varepsilon_0 + \chi_E$  is the so called dielectric permittivity of the background material,<sup>17</sup> which was considered almost independent of external electric field in linear regime.<sup>21</sup>

From the thermodynamic relation,  $dg = -\mathbf{D}(\mathbf{E}) \cdot d\mathbf{E}$  with  $\mathbf{D}$  to be a linear function of  $\mathbf{E}$ , the Gibbs free energy can be expressed as  $g = f(\mathbf{P})|_{\mathbf{E}=0} - \frac{1}{2} \mathbf{E} \cdot \mathbf{E} - \mathbf{P} \cdot \mathbf{E}$ , where the first term is the free energy without electric field. Since the epitaxial ultrathin film is fully constrained by the misfit strain from the substrate, the first term should be the Helmholtz free energy density with the consideration of internal elastic interaction. Without surface effect and external electric field, the Helmholtz free energy density of a constrained film can be written as<sup>23</sup>

$$f(\mathbf{P})|_{\mathbf{E}=0} = f(\mathbf{P})|_{\boldsymbol{\sigma}=0} + f_{\text{Ela}}, \quad (2)$$

where  $\mathbf{P}$  is the polarization,  $f(\mathbf{P})|_{\boldsymbol{\sigma}=0}$  is the free energy density of the unstressed film, and  $\boldsymbol{\sigma}$  is the stress tensor. The second term of Eq. (2) is the elastic energy density, which describes the interaction of the internal strain or stress with external stress. We only consider a single-domain *c* structure with the spontaneous polarization perpendicular to the film surface. The free energy density can be reduced to

$$f(P)|_{\mathbf{E}=0} = f(P)|_{\boldsymbol{\sigma}=0} + \frac{1}{2} \bar{G} (u_m - Q_{12} P^2)^2, \quad (3)$$

with the effective elastic modulus for a tetragonal crystal given by  $\bar{G} = C_{11} - C_{12} - 2C_{13}^2/C_{33} = 1/(s_{11} + s_{12})$ ;  $C_{ij}$  and  $s_{ij}$  are the elastic modulus and compliant coefficients of the film, respectively;  $u_m$  is the misfit strain induced by the substrate;  $Q_{12}$  is the electrostrictive coefficient. Using field-free uniform and infinite dimension crystal as the reference,  $F(P)|_{\boldsymbol{\sigma}=0}$  is the free energy that can be expressed in the form of a Landau free energy functional in terms of the order parameter  $P$ . The equilibrium system under an electrostatic field  $E$  normal to the film surface can be reached by minimizing the Gibbs free energy.<sup>23</sup> With the contributions of elastic energy, surface energy, and electrostatic energy, the total free energy density can be rewritten as

$$\Phi = \left( \frac{\alpha^*}{2} P^2 + \frac{\beta^*}{4} P^4 + \frac{\gamma}{6} P^6 + \bar{G} u_m^2 - \frac{1}{2} \varepsilon_d E^2 - PE \right) l + \Phi_S, \quad (4)$$

where  $\alpha^* = \alpha_0(T - T_{c0}) - 4Q_{13}u_m\bar{G}$ ,  $\beta^* = \beta + 4Q_{13}^2\bar{G}$  are the renormalized coefficients considering the misfit strain  $u_m$ ;  $\alpha_0$ ,  $\beta$ , and  $\gamma$  are the expansion coefficients of the Landau

free energy;  $T$  is the temperature;  $T_{c0}$  is the Curie temperature of the counterpart bulk material;  $s_{ij}$  is the elastic compliance tensor;  $\Phi_S$  is the surface energy, which may be written as a Taylor series expansion of  $P$ :  $\Phi_S = (\xi_1 - \xi_2)P + (\eta_1 + \eta_2)P^2/2$ , where  $\xi_1$ ,  $\eta_1$  and  $\xi_2$ ,  $\eta_2$  are expansion coefficients of the two surfaces, respectively.<sup>19</sup> Since the two electrodes are the same, the surface energy can be simplified as  $\Phi_S = \eta P^2$ . The electrostatic field  $E$  is actually the total electric field, i.e., the effective external electric field  $E_{\text{eff}}$  plus the depolarization field  $E_d$ . As shown in Fig. 1(b), the effective voltage on the film should consider the voltage drop in the electrodes because of the existence of an electric field inside the imperfect conductor electrodes.<sup>13</sup>

## B. Depolarization and effective electric field

The depolarization field due to imperfect conductor electrodes was believed to play an important role in the existence of the polarization in the thin films under short circuit condition.<sup>10,16,17</sup> The electric field distribution inside the electrode can be calculated by solving the Poisson's equation.<sup>16</sup> The one-dimensional Poisson's equation is given by

$$\frac{dD}{dx} = \rho(x), \quad (5)$$

where  $D = \varepsilon_e E_e + P_e$ ,  $E_e$  and  $\varepsilon_e$  are the electric field and the dielectric constant of the electrode, respectively. The spontaneous polarization  $P_e$  is zero inside the electrode, and  $\rho$  is the excess charge density. Assuming  $n_0$  and  $n$  are the charge densities without and with the electric field, respectively, the Poisson's equation can be expressed as

$$\varepsilon_e \frac{dE_e}{dx} = -q(n - n_0). \quad (6)$$

In the equilibrium state, electron distribution becomes statistically static in the electrode, so the total current density is equal to zero<sup>16</sup>

$$I_{\text{tot}} = 0 = qE_e(x)\mu_n n(x) + \mu_n n(x) \frac{d\xi}{dn} \frac{dn}{dx}, \quad (7)$$

with  $\frac{d\xi}{dn} = \frac{\pi^{4/3} \hbar^2 n^{-1/3}}{3^{1/3} m_{\text{eff}}} = K$ . Assuming  $n - n_0 \ll n_0$ , the following equation about the electric field can be derived as

$$E_e(x) = -\frac{\varepsilon_e K}{q^2} \frac{dn}{dx} = l_s^2 \frac{d^2 E_e(x)}{dx^2}, \quad (8)$$

with  $l_s = \sqrt{K\varepsilon_e}/q$ , known as the screening length that characterizes the free charge distribution in the electrode. Assuming the polarization induced compensation charges in the electrodes are  $\pm q_e$ , the boundary conditions are  $E_e(x) = 0$  at the outer surface of the electrode and  $E_e(x) = q_e/\varepsilon_e$  at the interface of the film and the electrode, the solution of Eq. (8) is given by

$$E_e(x) = \frac{q_e}{\varepsilon_e} \frac{\sinh[(l_e + x)/l_s]}{\sinh(l_e/l_s)}. \quad (9)$$

With the application of an external voltage  $V$ , the static electric balance condition can be written as<sup>24</sup>

$$2 \int_{l_e} E_e dx + \int_l E_f dx = V. \quad (10)$$

Using the displacement continuity at the interface:  $D_f = \epsilon_b E_f + P = q_e$ , the total electric field can be expressed as  $E_f = (q_e - P)/\epsilon_b$ . Submitting it into Eq. (10), one can derive the induced compensation charges  $q_e$

$$q_e = (P + V\epsilon_b/l)\theta, \quad (11)$$

where  $\theta = \frac{l}{\epsilon_b} / (\frac{2l}{\epsilon_e} + \frac{l}{\epsilon_b})$ . With the expression of  $q_e$ , the total effective electric field in the film can be written as

$$E_f = \frac{-P(1-\theta)}{\epsilon_b} + \frac{V}{l}\theta = E_d + E_{\text{eff}}. \quad (12)$$

The first term is the depolarization field  $E_d$  and the second term is the effective electric field in the film with the consideration of the potential drop in the electrodes. Obviously, if there is no external applied voltage, the internal electric field is just the depolarization field under short circuit condition, i.e.,  $E_f = E_d$ , which is also widely used in metal/ferroelectric/metal capacitors.<sup>16,17</sup> If the film is thick enough, or  $l_e$  is very small, or  $\epsilon_e$  is very large, then  $\theta \sim 1$ , the incomplete screening effect can be ignored resulting zero depolarization field under the short circuit condition. The internal electric field will be simply  $E = V/l$  under applied voltage  $V$ . However, when the film is ultrathin and the electrodes are not perfect conductors, both the depolarization field and the effective electric field in the film will be affected by the trapped charges inside the electrodes, which caused incomplete screening effect.

### C. Physical properties

Under the total internal electric field  $E_d + E_{\text{eff}}$ , the equation governing the time-dependence of polarization can be derived by the variation of the total free energy in Eq. (4) with respect to the polarization

$$\frac{\partial P}{\partial t} = -M \frac{\delta \Phi}{\delta P} = -Ml[(\alpha^{**}P + \beta^*P^3 + \gamma P^5) - E'_{\text{eff}}], \quad (13)$$

where  $\alpha^{**} = \alpha^* + (1 - \theta^2)\epsilon_f^{-1} + 2\eta l^{-1}$  is the renormalized coefficient,  $M$  is the kinetic coefficient, the last term is the modified effective electric field  $E'_{\text{eff}} = \theta^2 V/l$ .

The susceptibility related to the spontaneous polarization  $\chi_S$  under zero external field can be expressed via the total free energy as

$$\chi_S = \frac{\partial P}{\partial E'_{\text{eff}}} \approx \left( \frac{\partial^2 \Phi}{\partial P^2} \right)^{-1} = (\alpha^{**} + 3\beta^*P^2 + 5\gamma P^4)^{-1}. \quad (14)$$

The spontaneous susceptibility  $\chi_S$  is usually much larger than the linear field-dependent susceptibility  $\chi_E$ , especially near the transition point. However,  $\chi_E$  will become comparable when the thickness of the film is very small or when the spontaneous susceptibility is small. We should note that  $\chi_S$  here is taken under a non-zero depolarization field and with surface discontinuity.

Nonlinear piezoelectric effect may exist in ultrathin film, because the field could be very high even the applied voltage may be low.<sup>25</sup> Under the boundary condition of uniform in-plane strain  $S_1 = S_2 = u_m$  and vertical stress free  $T_3 = 0$ , the total elastic strain in the vertical direction can be derived as

$$S_3 = \frac{2s_{13}u_m}{s_{11} + s_{12}} + \left( Q_{11} - \frac{2s_{13}Q_{12}}{s_{11} + s_{12}} \right) (P_E + P)^2 = \tilde{s}u_m + \tilde{Q}(P_E + P)^2, \quad (15)$$

where  $\tilde{s}$  and  $\tilde{Q}$  are the effective elastic compliance ratio and electrostrictive coefficient, respectively. The first term of Eq. (15) is the misfit strain induced by the substrate. The effective piezoelectric coefficients can be approximated as

$$d_{33}^* = \frac{dS_3}{dE_{\text{eff}}} = 2\tilde{Q}(P + P_E) \frac{(\partial P_E + P)}{dE_{\text{eff}}} = 2\tilde{Q}(P + \chi_E E_f)(\chi_E^* + \theta\chi_S), \quad (16)$$

where  $\chi_E^* = \partial P_E / \partial E_{\text{eff}} = \partial \chi_E E_f / \partial E_{\text{eff}} = \chi_E (1 - \frac{1-\theta}{\epsilon_b} \theta \chi_S)$  and  $\partial P / \partial E_{\text{eff}} = \theta \chi_S$ . Because of the rigid clamping by the substrate, there is no induced in-plane strain in the film, so that the effective  $d_{31}^*$  should always be zero.

### III. RESULTS AND DISCUSSIONS

The parameters used for  $\text{PbTiO}_3$  and electrode  $\text{SrRuO}_3$  were taken from Ref. 10. The calculations were carried out at room temperature for films with the thickness less than 6 nm. The misfit strain induced by the substrate is taken as  $-2.6\%$ . Under short circuit condition with imperfect conductor electrodes, polarization disappears below a critical thickness. For  $\text{PbTiO}_3$  film with an applied strain of  $-2.6\%$ , the critical thickness is calculated to be around 24 Å at room temperature (Fig. 2). This value is higher than the reported 0.8 nm detected by using synchrotron radiation<sup>8</sup> due to the consideration of significant depolarization effect induced by imperfect electrodes. Lower temperature, better compatible oxide-oxide interface, and larger compressive strain can counterbalance the degradation of the polarization induced by size reduction.

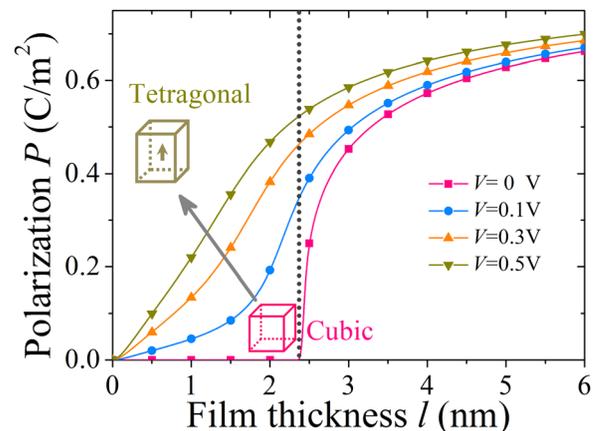


FIG. 2. Polarization vs. film thickness under an applied voltage.

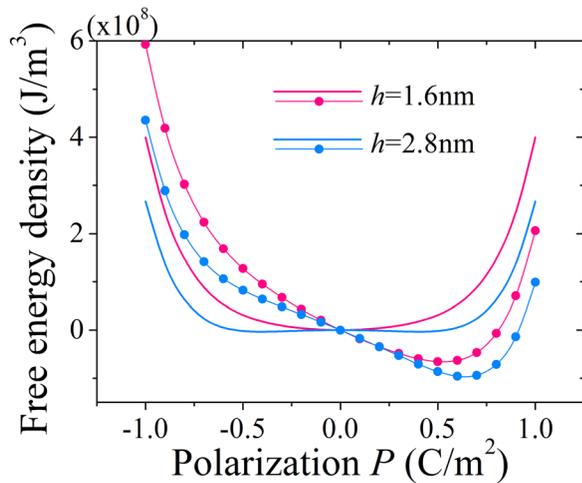


FIG. 3. Energy density vs. polarization without (solid line) and with (dotted solid line) external voltage for films with thicknesses below and above the critical thickness.

Under an applied voltage, a finite spontaneous polarization can exist below the critical thickness. The discontinuity of polarization with respect to film thickness is changed from a step-type to a continuous one (Fig. 2), which can be understood from the free energy chart. As shown in Fig. 3, the free energy density without electric field for thickness lower than the critical thickness has a minimum at zero polarization. With the application of a finite voltage, the energy becomes asymmetric and the minimum moves to a non-zero polarization. However, these potential wells for ultrathin films are much shallower than those of thick films, so that the system will have higher responsiveness to external stimuli.

The effective external electric field and depolarization field are important for many physical phenomena, such as dielectric breakdown, fatigue, interfacial potential, etc. As shown in Fig. 4, the total field contains two parts, without applied voltage,  $E_f$  vanishes as the depolarization becomes zero below the critical thickness, and has a linear relation with the polarization, but an inverse relation with film thickness as given in Eq. (12). Under an external voltage of 0.5 V,

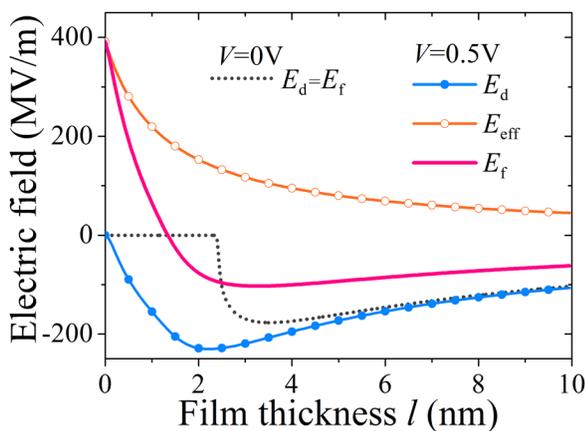


FIG. 4. Effective external electric field, depolarization field, and the total electric field in the film. Dashed line is for zero voltage and solid lines are for voltage of 0.5 V.

the effective internal field is negative even under a positive voltage due to the large depolarization field for film with thickness larger than the critical thickness. This negative total electric field will result a negative field-induced polarization  $P_E$ , although the spontaneous polarization  $P$  is still positive in the field direction.

Dielectric property correlates directly with piezoelectric behavior in ferroelectric materials. The dielectric constant has a sharp increase in the vicinity of the critical point and could diverge at the critical point. The temperature dependence of the dielectric susceptibility can be approximated by Eq. (14). As shown in Fig. 5, the dielectric susceptibility shows a sharp increase in the vicinity of the critical thickness. However, with the application of a small voltage, the singularity at the critical point becomes a finite peak with reduced amplitude, and the dielectric peak is broadened and shifted towards smaller thickness.

Piezoelectric constant usually decreases with the reduction of film thickness and becomes zero below the critical thickness due to the disappearance of the polarization. With the increase of voltage, the critical thickness seems disappear corresponding with a relatively large induced-polarization below the critical thickness. According to the relationship between the dielectric constant and piezoelectric coefficient, the piezoelectric coefficient will have a sharp increase in the vicinity of critical thickness, showing a sharp dielectric peak. Below the critical thickness, the polarization and piezoelectricity is dependent on the amplitude of the applied voltage. As shown in Fig. 6, the effective piezoelectric coefficient increases with the increase of film thickness and may have a small cusp in the vicinity of critical thickness, then, monotonically increases with the increase of film thickness. We should note that the calculated effective piezoelectric coefficient is smaller than  $d_{33} = 2\tilde{Q}P\chi_S$  as shown in the inset of Fig. 6 due to the depolarization effect. Even though, the value predicted in this work is still one order of magnitude higher than the reported experiment value of 2-5 pm/V,<sup>8</sup> which leaves much room for the improvement of film quality to produce higher piezoresponse.

The piezoresponse under a small applied voltage in ultrathin ferroelectric film is usually nonlinear. As shown in

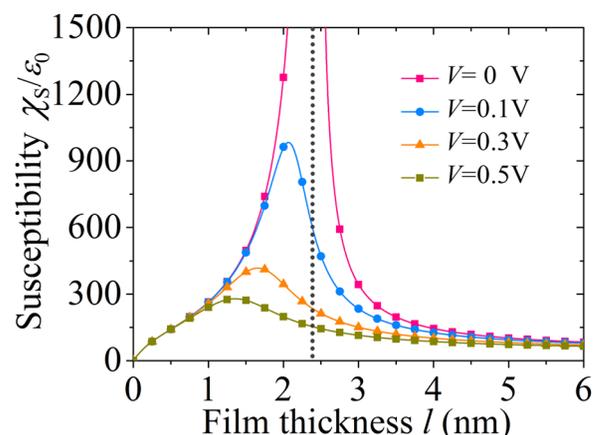


FIG. 5. Relative dielectric susceptibility  $\chi_S/\epsilon_0$  vs. film thickness without and with applied voltage.

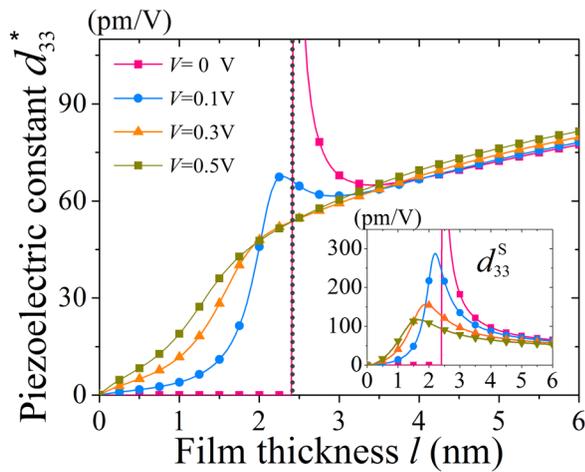


FIG. 6. Effective piezoelectric coefficient  $d_{33}^*$  vs. film thickness. Inset is the theoretical value calculated by  $d_{33}^s = 2\hat{Q}P\chi_3^s$  for comparison.

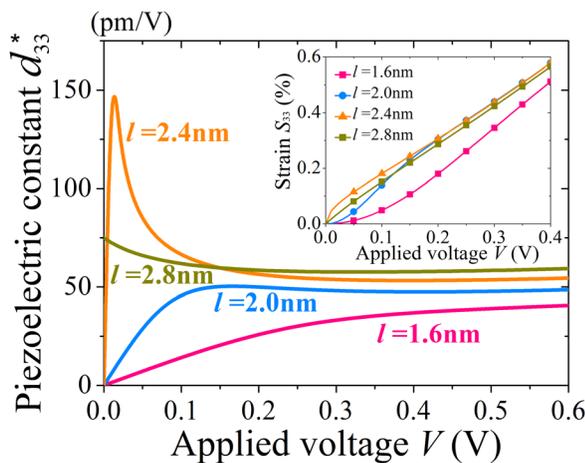


FIG. 7. Piezoresponse under a small voltage for nano scale films with different thicknesses. Inset is the piezoelectric strain under small voltage.

Fig. 7, the effective piezoelectric constant vs. applied voltage shows a very large diversity for films of different thicknesses. When the film thickness is below the critical thickness, the effective piezoelectric constant almost linearly increases with the applied voltage until the voltage reached the critical value, then shows some reduction and gradually approaches a constant. For films with thickness above the critical thickness, the effective piezoelectric constant will decrease with the applied voltage and gradually approaches a constant. The critical voltage can be derived by setting the derivative of  $d_{33}^*$  with respect to applied voltage  $V$  to zero. For any given film thickness, the field induced piezoelectric strain is almost linear as shown in the inset of Fig. 7.

#### IV. SUMMARY AND CONCLUSIONS

Physical properties of ultrathin ferroelectric films under an external voltage were theoretically studied with the

consideration of the depolarization field and the voltage drop inside the imperfect electrodes. Polarization in an ultrathin film under an applied field can persist below the critical thickness and the behavior is quite different from that of the bulk. The nearly first order like thickness driven phase transition behavior has been smeared out due to the existence of polarization below the critical thickness with applied voltage. In particular, the singularities of the dielectric and piezoelectric constants at the critical thickness were all broadened by the applied voltage with reduced amplitude. Although the piezoelectric constant shows large electric field dependence and is quite nonlinear, but the piezoelectric strain for a given film thickness still behaves almost linear with respect to the applied voltage.

#### ACKNOWLEDGMENTS

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