Dielectric and piezoelectric properties of (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin films*

Jian-Hua Qiu(邱建华)[1,2,†], Zhi-Hui Chen(陈智慧)[1,2], Xiu-Qin Wang(王秀琴)[1,2], Ning-Yi Yuan(袁宁一)[1,2], and Jian-Ning Ding(丁建宁)[1,2]

[1] *Jiangsu Province Cultivation Base for State Key Laboratory of Photovoltaic Science and Technology, Changzhou University, Changzhou* 213164, *China*
[2] *Jiangsu Collaborative Innovation Center of Photovolatic Science and Engineering, Changzhou University, Changzhou* 213164, *China*

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A phenomenological Landau–Devonshire theory is developed to investigate the ferroelectric, dielectric, and piezoelectric properties of (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films. At room temperature, the tetragonal a_1 phase, the orthorhombic a_2c phase, the triclinic γ_1 phase, and the triclinic γ_2 phase are stable. The appearance of the negative polarization component P_2 in the a_2c phase and the γ_1 phase is attributed to the nonlinear coupling terms in the thermodynamic potential. The γ phase of the $Pb(Zr_{1-x}Ti_x)O_3$ thin films has better dielectric and piezoelectric properties than the a_2c phase and the a_1 phase. The largest dielectric and piezoelectric coefficients are obtained in the $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film. The piezoelectric coefficient of 110–150 pm/V is obtained in the (110) oriented $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film, and the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film has the remnant polarization and relative dielectric constant of 50 μC/cm^2 and 100, respectively, which are in agreement with the experimental measurements reported in the literature.

Keywords: polarization, dielectric property, piezoelectric property

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1. Introduction

The $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) thin films have attracted a great deal of attention for use in nonvolatile ferroelectric random access memories, sensors, and actuators because of their superior properties of large spontaneous polarization and piezoelectricity.[1–6] Some researches focused on the (001) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin films grown on compressive cubic substrates with large remanent polarization,[7–9] such as $SrTiO_3$. However, their dielectric and piezoelectric properties were not so prominent due to the residual strain in the $Pb(Zr_{1-x}Ti_x)O_3$ thin films.[8] Actually, the dielectric and piezoelectric properties are strongly affected by the film orientation which is related to the substrates or the buffer layers,[10–12] and good dielectric and piezoelectric properties were observed in the (110) oriented films.[13–15]

Experimentally, the effect of the buffer layer on the film orientation and the piezoelectric properties was investigated. Epitaxial $Pb(Zr_{0.52}Ti_{0.48})O_3$ thin films with and without a CeO_2 buffer layer were deposited on silicon substrates respectively using pulsed laser deposition.[14] The (110) oriented film was obtained on a YSZ/Si substrate, while the (001) oriented film was fabricated on a CeO_2/YSZ/Si substrate. The longitudinal piezoelectric coefficient d_{33} of the (110) oriented film was larger than that of the (001) oriented film. On the other hand, the effect of the substrate on the film orientation and the dielectric properties in epitaxial $Pb(Zr_{1-x}Ti_x)O_3$ thin films was investigated for different Ti concentrations.[15] The dielectric constant in the (110) oriented film was larger than that in the (001) oriented film, and the $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin films had the better dielectric property.

Although the experimental results indicated that the (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin films have better dielectric and piezoelectric properties than the (001) oriented films, there are no theoretical reports on the (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin films. As we known, the Landau–Devonshire theory is an effective method to investigate the ferroelectric, dielectric, and piezoelectric properties of conventional ferroelectrics.[16–20] Therefore, (110) or (111) oriented films can be analyzed theoretically by the transformations of polarization, stress, and electric field in the frame of the Landau–Devonshire theory.[21,22]

Thus, the goal of this paper is to investigate the ferroelectric, dielectric, and piezoelectric properties of (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films. The effects of composition and misfit strain on those properties are evaluated. New ferroelectric phases, which are rare in the (001) oriented films, result in the excellent dielectric and piezoelectric properties.

2. Model and method

We consider a (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin film grown on a thick substrate. The elastic Gibbs function G of a cubic ferroelectric is given by the Landau–Devonshire theory[22,23]

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†Corresponding author. E-mail: jhqiu@cczu.edu.cn

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$$
\begin{aligned}
G = {} & \alpha_1(\tilde{P}_1^2+\tilde{P}_2^2+\tilde{P}_3^2)+\alpha_{11}(\tilde{P}_1^4+\tilde{P}_2^4+\tilde{P}_3^4)\\
& +\alpha_{12}(\tilde{P}_1^2\tilde{P}_2^2+\tilde{P}_1^2\tilde{P}_3^2+\tilde{P}_2^2\tilde{P}_3^2)\\
& +\alpha_{111}(\tilde{P}_1^6+\tilde{P}_2^6+\tilde{P}_3^6)+\alpha_{112}[\tilde{P}_1^4(\tilde{P}_2^2+\tilde{P}_3^2)\\
& +\tilde{P}_2^4(\tilde{P}_1^2+\tilde{P}_3^2)+\tilde{P}_3^4(\tilde{P}_1^2+\tilde{P}_2^2)]\\
& +\alpha_{123}\tilde{P}_1^2\tilde{P}_2^2\tilde{P}_3^2-\frac{1}{2}s_{11}(\tilde{\sigma}_1^2+\tilde{\sigma}_2^2+\tilde{\sigma}_3^2)\\
& -s_{12}(\tilde{\sigma}_1\tilde{\sigma}_2+\tilde{\sigma}_1\tilde{\sigma}_3+\tilde{\sigma}_2\tilde{\sigma}_3)\\
& -\frac{1}{2}s_{44}(\tilde{\sigma}_4^2+\tilde{\sigma}_5^2+\tilde{\sigma}_6^2)-Q_{11}(\tilde{\sigma}_1\tilde{P}_1^2+\tilde{\sigma}_2\tilde{P}_2^2+\tilde{\sigma}_3\tilde{P}_3^2)\\
& -Q_{12}[\tilde{\sigma}_1(\tilde{P}_2^2+\tilde{P}_3^2)+\tilde{\sigma}_2(\tilde{P}_1^2+\tilde{P}_3^2)+\tilde{\sigma}_3(\tilde{P}_1^2+\tilde{P}_2^2)]\\
& -Q_{44}(\tilde{P}_2\tilde{P}_3\tilde{\sigma}_4+\tilde{P}_1\tilde{P}_3\tilde{\sigma}_5+\tilde{P}_1\tilde{P}_2\tilde{\sigma}_6)\\
& -\tilde{E}_1\tilde{P}_1-\tilde{E}_2\tilde{P}_2-\tilde{E}_3\tilde{P}_3, \qquad (1)
\end{aligned}
$$

where $\tilde{P}_i$, $\tilde{\sigma}_i$, and $\tilde{E}_i$ are the polarization, stress, and electric field components defined in the crystallographic reference frame ($\tilde{X}_1$, $\tilde{X}_2$, $\tilde{X}_3$) aligned with the cube axes of the material; α_1, $\alpha_{i,j}$, and $\alpha_{i,j,k}$ are the dielectric stiffness coefficients; s_{ij} are the elastic compliances at constant polarization; and Q_{ij} are the electrostrictive coefficients. These parameters of the $Pb(Zr_{1-x}Ti_x)O_3$ thin film used for calculations come from Ref. [24]. In order to construct the thermodynamic potential in the film frame (X_1, X_2, X_3), the polarization $\tilde{P}_i$, stress $\tilde{\sigma}_i$, and electric field $\tilde{E}_i$ are transformed by using the frame transformation $\tilde{X}_i = A_{i,j}X_j$, where $A_{i,j}$ is the frame transformation matrix. For a (110) oriented film, where the X_3 axis is parallel to [011] and the X_1 axis is parallel to [100], $A_{i,j}$ is given by

$$
A_{i,j} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1/\sqrt{2} & -1/\sqrt{2} \\ 0 & 1/\sqrt{2} & 1/\sqrt{2} \end{pmatrix}. \qquad (2)
$$

Based on the previous work,[16] the thermodynamic potential of a single domain film is rewritten as

$$
G_{\text{film}} = G + S_1\sigma_1 + S_2\sigma_2 + S_6\sigma_6. \qquad (3)
$$

The mechanical boundary conditions are given by $S_1 = S_2 = S_{\text{m}}$, where S_{m} is the misfit strain along the in-plane direction, $\sigma_3 = \sigma_4 = \sigma_5 = 0$, and $S_6 = 0$. All the stress components in Eq. (3) can be eliminated by the mechanical boundary conditions and $\partial G/\partial\sigma_1 = \partial G/\partial\sigma_2 = -S_{\text{m}}$, $\partial G/\partial\sigma_6 = 0$. Therefore, the thermodynamic potential G_{film} is expanded as a function of polarization P_i, temperature T, misfit strain S_{m}, and electric field E_i

$$
\begin{aligned}
G_{\text{film}} = {} & \alpha_1^*P_1^2+\alpha_2^*P_2^2+\alpha_3^*P_3^2+\alpha_{11}^*P_1^4+\alpha_{22}^*P_2^4\\
& +\alpha_{33}^*P_3^4+\alpha_{12}^*P_1^2P_2^2+\alpha_{13}^*P_1^2P_3^2\\
& +\alpha_{23}^*P_2^2P_3^2+\alpha_{2223}(P_2P_3^3+P_2^3P_3)\\
& +\alpha_{1123}P_1^2P_2P_3+\alpha_{111}^*P_1^6+\alpha_{333}^*(P_2^6+P_3^6)\\
& +\alpha_{112}^*(P_1^4P_2^2+P_1^4P_3^2)+\alpha_{122}^*(P_1^2P_2^4+P_1^2P_3^4)\\
& +\alpha_{223}^*(P_2^4P_3^2+P_2^2P_3^4)\\
& +\alpha_{123}^*P_1^2P_2^2P_3^2-E_1P_1-E_2P_2-E_3P_3. \qquad (4)
\end{aligned}
$$

The polarization and its dependence on the misfit strain are given by the equilibrium condition $\partial G_{\text{film}}/\partial P_i = 0$ ($i = 1,2,3$). For investigating the dielectric property of the $Pb(Zr_{1-x}Ti_x)O_3$ thin film, we can derive an explicit expression for the reciprocal dielectric susceptibilities by differentiating the thermodynamic potential G_{film}, i.e., $\chi_{ij} = \partial^2 G_{\text{film}}/\partial P_i\partial P_j$. Consequently, the matrix (χ) of the reciprocal dielectric susceptibilities is obtained as

$$
\chi = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{12} & \chi_{22} & \chi_{23} \\ \chi_{13} & \chi_{23} & \chi_{33} \end{pmatrix}. \qquad (5)
$$

The matrix inversion then enables us to find the dielectric susceptibility $\eta = \chi^{-1}$ and dielectric constant $\varepsilon_{ij} = \varepsilon_0 + \eta_{ij}$, here ε_0 is the dielectric constant of free space. By substituting the equilibrium values P_i of the polarization components into the expression derived for χ_{ij} via Eq. (4), the small-signal dielectric response ε_{ij} is calculated. Generally speaking, the piezoelectric coefficient d_{ln} is calculated as $d_{ln} = \partial S_n/\partial E_l = b_{kn}\eta_{kl}$, where $b_{kn} = \partial S_n/\partial P_k$ and η_{kl} is the dielectric susceptibility. The strain can be obtained by differentiating the elastic Gibbs function G, $S_n = -\partial G/\partial\sigma_n$.[25]

3. Results and discussion

For investigating the dielectric and piezoelectric properties of the (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films, the temperature–misfit strain phase diagrams are constructed firstly.[26] At room temperature, only the orthorhombic a_2c phase ($P_1 = 0$, $P_2 < 0$, $P_3 > 0$), the tetragonal a_1 phase ($P_1 > 0, P_2 = P_3 = 0$), the triclinic γ_1 phase ($P_1 > 0$, $P_2 < 0$, $P_3 > 0$), and the triclinic γ_2 phase ($P_1 > 0$, $P_2 > 0$, $P_3 > 0$) are stable. The effect of the misfit strain on the polarization at room temperature is presented in Fig. 1. The $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, and 0.6) thin films experience the first order phase transition from the orthorhombic a_2c phase to the triclinic γ phase, and the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film has the first order phase transition from the orthorhombic a_2c phase to the tetragonal a_1 phase. Even in the γ phase, the first order γ_1–γ_2 phase transition may take place in the $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$ and 0.5) thin films. Therefore, all the polarization components P_i are discontinuous. The polarization components P_2 and P_3 of the orthorhombic a_2c phase are negative and positive, respectively. Moreover, the polarization component P_3 increases and P_2 decreases with increasing compressive misfit strain, because the $Pb(Zr_{1-x}Ti_x)O_3$ thin films are grown along the [011] direction and the compressive misfit strain is beneficial to induce the polarization component P_3. The appearance of the negative polarization component P_2 in the a_2c phase and the γ_1 phase is attributed to the nonlinear coupling terms $\alpha_{2223}(P_2P_3^3+P_2^3P_3)$ and $\alpha_{1123}P_1^2P_2P_3$ in Eq. (4). Experimentally, the $Pb(Zr_{0.35}Ti_{0.65})O_3$,[11] $Pb(Zr_{0.42}Ti_{0.58})O_3$,[12] and $Pb(Zr_{0.26}Ti_{0.74})O_3$[15] thin films had the remanent polarizations P_r of 50 μC/cm^2, 45 μC/cm^2, and 48 μC/cm^2, respectively, which are in agreement with the theoretical results.

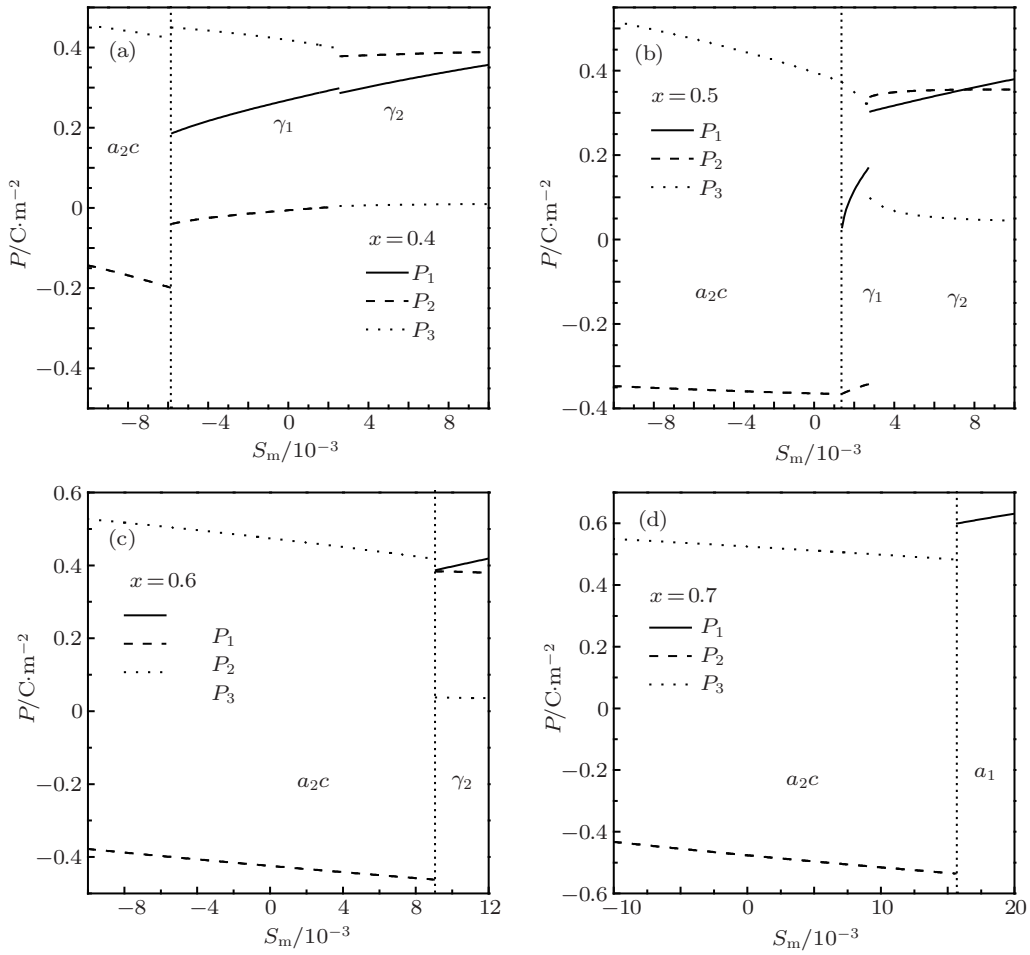


Fig. 1. The effect of the misfit strain on the polarization components P_i in $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films at room temperature.

The misfit strain dependence of the spontaneous polarization P_s in the (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ thin films is shown in Fig. 2. The spontaneous polarizations of the orthorhombic a_2c phase and the triclinic γ phase have magnitudes of $P_s = \sqrt{P_2^2 + P_3^2}$ and $P_s = \sqrt{P_1^2 + P_2^2 + P_3^2}$, respectively. With the increase of the compressive misfit strain, the spontaneous polarization P_s of the orthorhombic a_2c phase increases for the Ti composition of $x \leq 0.6$, while it decreases for the Ti composition of 0.7. This is because the spontaneous polarization P_s of the a_2c phase mainly depends on the polarization component P_3 and P_2 for $x \leq 0.6$ and $x = 0.7$, respectively. For the $Pb(Zr_{0.6}Ti_{0.4})O_3$ thin film, the spontaneous polarization P_s of the triclinic γ_1 phase firstly increases and then decreases with the variation of the misfit strain from compressive strain to tensile strain. Moreover, the polarization P_s of the triclinic γ_2 phase increases with further increase of the tensile misfit strain. The spontaneous polarization P_s in the $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film firstly decreases in the triclinic γ_1 phase and then increases in the triclinic γ_2 phase with increasing tensile misfit strain. The spontaneous polarizations P_s of the triclinic γ_2 phase in the $Pb(Zr_{0.4}Ti_{0.6})O_3$ thin film and the tetragonal a_1 phase in the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film both increase as the tensile misfit strain increases. As is seen from the figure, the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film has the largest spontaneous polarization due to its large Ti composition.

The dependence of the relative dielectric constant $\varepsilon_{ii}/\varepsilon_0$ on the misfit strain is plotted in Fig. 3. Because of the first order ferroelectric–ferroelectric phase transitions at room temperature, all the relative dielectric constants are discontinuous. In the a_2c phase, the relative dielectric constants $\varepsilon_{11}/\varepsilon_0$ and $\varepsilon_{33}/\varepsilon_0$ decrease with increasing compressive misfit strain due to the increase of the polarization component P_3, and the decrease of the polarization component P_2 results in the increase of the relative dielectric constant $\varepsilon_{22}/\varepsilon_0$. In addition, the increase of the tensile misfit strain decreases the relative dielectric constants $\varepsilon_{ii}/\varepsilon_0$ of the a_1 phase for the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film because of the increase of the polarization component P_1. The $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film has the best dielectric property with a relative dielectric constant $\varepsilon_{33}/\varepsilon_0$ of 4000 at the misfit strain of 3×10^{-3}, which has potential applications in semiconductor memory devices. This is because the Ti composition of the $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film is near the morphotropic phase boundary (MPB) and the large dielectric property is obtained, which is in agreement with the experimental result. The $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film has the smallest dielectric constant due to its large spontaneous polarization, and the magnitude of 100 is in agreement with the experimental measurement.[15]

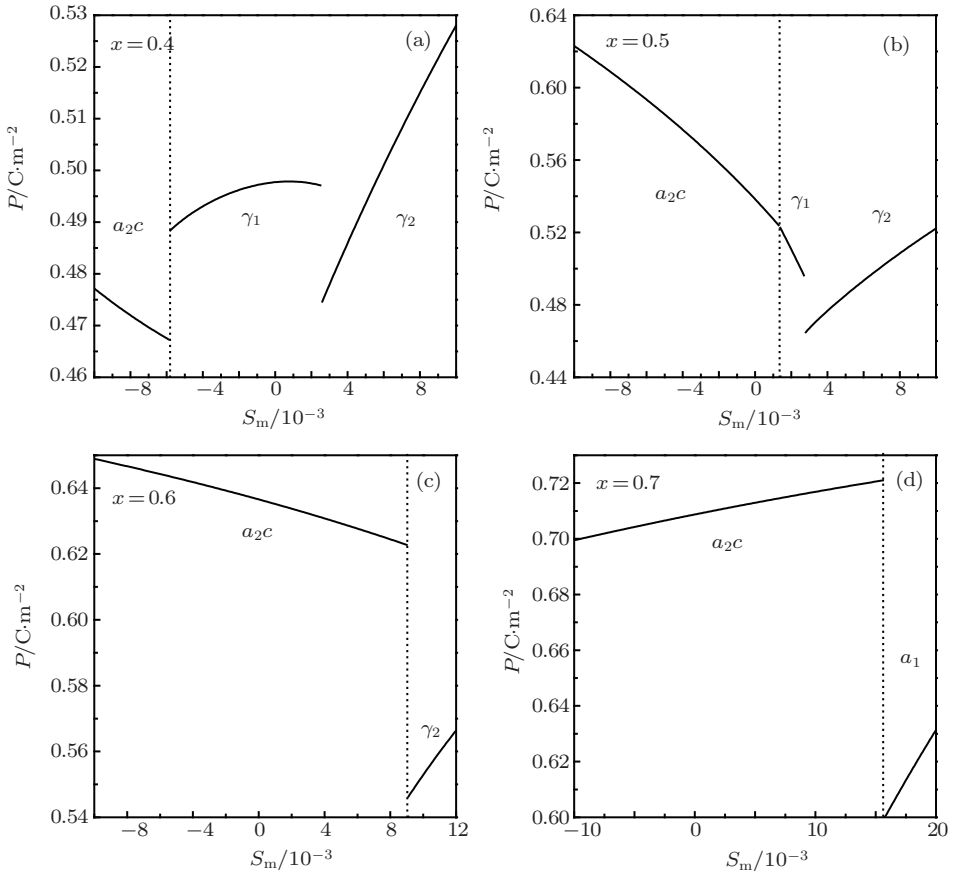


Fig. 2. The room temperature spontaneous polarizations P_s of $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films as a function of the misfit strain.

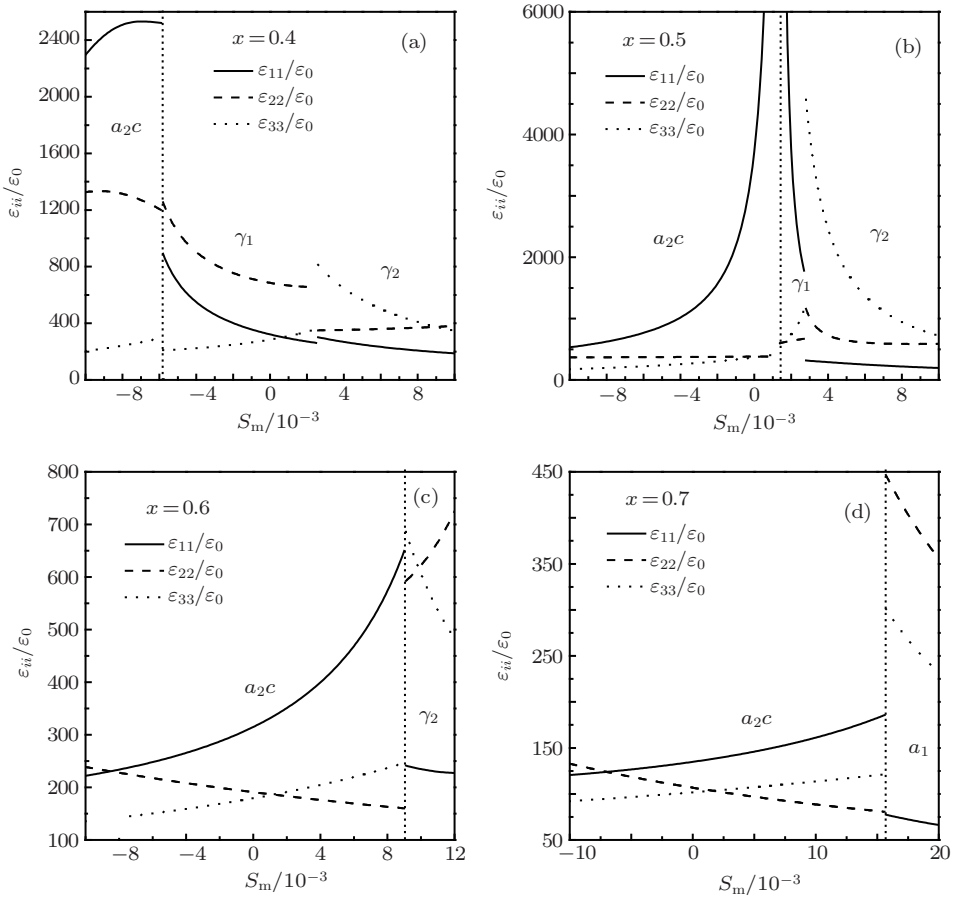


Fig. 3. The dependence of relative dielectric constants $\varepsilon_{ii}/\varepsilon_0$ on the misfit strain in $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films.

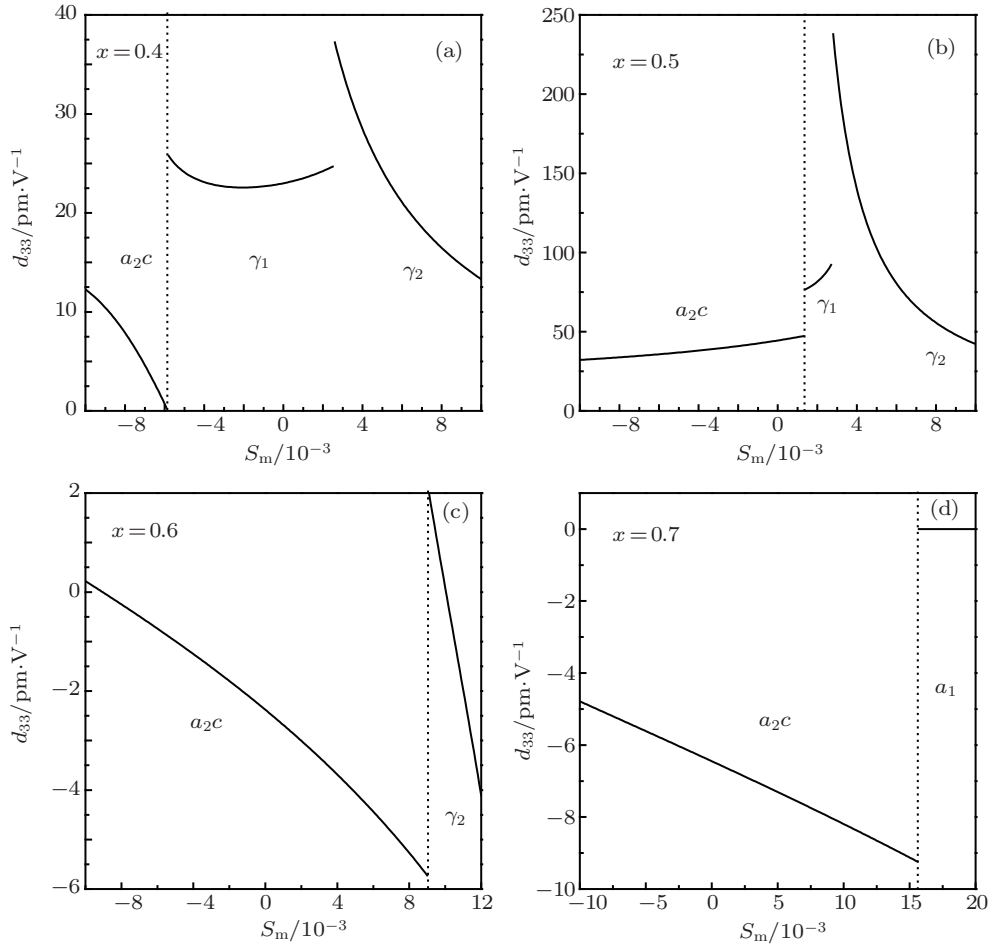


Fig. 4. The piezoelectric coefficients of the $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films.

The dependence of the longitudinal piezoelectric coefficient d_{33} on the misfit strain for the $Pb(Zr_{1-x}Ti_x)O_3$ thin films is shown in Fig. 4. The piezoelectric coefficient of the a_2c phase deceases with the increase of the compressive misfit strain except for the $Pb(Zr_{0.6}Ti_{0.4})O_3$ thin film. Moreover, the piezoelectric coefficient of the a_1 phase is zero because the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film only has the in-plane polarization component P_1. The new ferroelectric γ phase results in the excellent piezoelectric property, which is larger than that of the a_2c phase and the a_1 phase. The $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film has the largest piezoelectric coefficient due to the existence of the MPB. Nguyen *et al.* reported that the (110) oriented $Pb(Zr_{0.52}Ti_{0.48})O_3$ thin film grown on $SrRuO_3$/YSZ/Si substrate has a longitudinal piezoelectric coefficient of 50–150 pm/V with the film thickness ranging from 100 nm to 2000 nm,[14,27] which is in good agreement with the theoretical result.

4. Conclusion

The ferroelectric, dielectric, and piezoelectric properties of the (110) oriented $Pb(Zr_{1-x}Ti_x)O_3$ ($x = 0.4$, 0.5, 0.6, and 0.7) thin films are investigated by the phenomenological Landau–Devonshire theory. The excellent dielectric and piezoelectric properties are obtained due to the new ferroelectric phases, such as the triclinic γ phase and the orthorhombic a_2c phase, which cannot be obtained in the (001) oriented films. The appearance of the negative polarization component P_2 in the a_2c phase and the γ_1 phase is attributed to the nonlinear coupling terms $\alpha_{2223}(P_2P_3^3 + P_2^3P_3)$ and $\alpha_{1123}P_1^2P_2P_3$. The $Pb(Zr_{0.5}Ti_{0.5})O_3$ thin film with the Ti composition around the morphotropic phase boundary has the largest dielectric and piezoelectric coefficients. Moreover, the $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin film has the remnant polarization and dielectric constant of 50 μC/cm^2 and 100, respectively, which are in accordance with the experimental measurements.

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