Robust ferroelectricity enhancement of PZT thin films by a homogeneous seed layer

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ABSTRACT

With its excellent ferroelectric properties such as large dielectric constant and large remanent polarization, PZT thin films are extensively used in micro-sensors and other devices. In this study, the sol–gel process was used to fabricate Pb(Zr0.52Ti0.48)O3 thin films with Pb(Zr0.52Ti1−x)O3 seed islands. The experimental consequences demonstrate that all the Pb(Zr0.52Ti0.48)O3 thin films with Pb(Zr0.52Ti1−x)O3 seeds show pure perovskite phase with no other impurity phases, and the electrical properties of Pb(Zr0.52Ti0.48)O3 thin films modified by Pb(Zr0.52Ti1−x)O3 seed islands with different Zr/Ti ratios are improved, such as remanent polarization increased, dielectric properties increased, coercive electric field decreased, leakage current density decreased. In particular, the electrical properties of the Pb(Zr0.52Ti0.48)O3 thin films with Pb(Zr0.52Ti1−x)O3 seed islands are the most optimal when the x is 0.52. This paper provides a new technique for optimizing the electrical properties of PZT thin films, which is of great significance for breaking through the bottleneck of the development of ferroelectric memory.
Introduction

The perovskite lead zirconate titanate Pb(Zr,Ti)O₃ (PZT) thin films are extensively used as micro-sensors, piezoelectric actuators, micro-electromechanical system devices, and nonvolatile ferroelectric random access memories (FeRAM), because of their large dielectric constant, excellent piezoelectric effects, and large remanent polarization [1–6]. Unfortunately, there are still some problems that need to be solved urgently for ferroelectric thin films, such as excessive leakage current density and poor fatigue resistance, which enormously affect the performance potential and stability of devices for FeRAM. Therefore, how to improve the performance of PZT thin films to meet the development needs of a new high-performance ferroelectric memory is a problem that we need to solve now [7–9].

In the past period, a considerable amount of research was carried out toward saving the above problems. Lu et al. [10] added ethylene glycol as solvent based on the original solvent, which contributed to the formation of nuclei and improved the electrical properties of PZT films. In order to control the phase composition of the PZT film to obtain more stable ferroelectric properties, Choi et al. [11] studied the effect of the O₂ gas ratio on the formation of the perovskite phase and conductivity of the PZT thin film by changing the O₂ gas ratio in the sputtering gas. Sun et al. [12] fabricated Mn-doped lead zirconate titanate thin films (PMZT) and Nd-doped lead zirconate titanate thin films (PNZT) by sol–gel process, respectively, and systematically research the effects of Mn and Nb doping on the domain wall mobility and oxygen vacancy concentration of PZT films. And also, Wu et al. [13] used Ba(Mg₁/₃Ta₂/₃)O₃ as the buffer layer to enhance the fatigue resistance of PZT thin films. Although certain properties of the PZT films can be improved to a certain extent, due to the significant heterogeneous interface between different materials, the PZT films are enslaved to diffusion and dead layer problems, which makes the films possess obvious defects. This increases the leakage current density, resulting in attenuation of the other electrical properties of PZT.
thin films [14–16]. Therefore, we propose a new method that selects PZT material itself as seed islands to control the growth of PZT thin films. Compared with the past methods, the first advantage of the method proposed in this paper is that the addition of the seed islands provides nucleation points for the growth of the film. Controlling the growth of the film interface through the seed islands can reduce the activation energy of the film nucleation growth, which is conducive to the rapid growth of the films, while reducing its annealing temperature and improving the compatibility with the CMOS process. We learned that some previous researches were based on the above-mentioned seed islands principle, by adding heterogeneous materials such as PbTiO3 [17] or SrTiO3 [18] as the seed islands to improve the performance of PZT thin films, but they still ignored the influence of diffusion and dead layer brought by heterogeneous interface on the microstructure and electrical properties of the films. So another advantage of our method is that since the composition of the seed islands is consistent with the film, based on the role of the seed islands, it reduces the defects and lattice mismatch caused by choosing other different materials as the seed islands to form a heterogeneous interface. The interface induction of seed islands ensures the formation of a high-quality PZT film consistent with the orientation of the seed islands during the subsequent PZT film formation process.

In this work, (111)-PZT thin films with different Zr/Ti ratios seed islands were grown by sol–gel [19, 20]. First of all, we added Pb(Zr$_{x}$Ti$_{1-x}$)O$_3$ seeds of different Zr/Ti ratios to the substrates. Then, we fabricated Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin films with the same thickness on these substrates [21]. Finally, the microstructure and electrical properties of these PZT films, such as remanent polarization, dielectric constant, leakage current density, were measured by various experimental instruments [22]. By comparing the measured experimental parameters, we systematically discussed the influence of Pb(Zr$_{x}$Ti$_{1-x}$)O$_3$ seed islands on the electrical properties of Pb(Zr$_{0.52}$-Ti$_{0.48}$)O$_3$ thin films. We found that PZT seed islands with different Zr/Ti ratios can commendably adjust the microstructure of the thin films. Moreover, the electrical properties of PZT ferroelectric thin films can be effectively enhanced by the seed islands. When the Zr/Ti ratio of seed islands was 52/48, the regulation effect was the most remarkable.

**Materials and methods**

**Materials and fabrication**

Here, we used the sol–gel method to fabricate PZT thin films in oxygen atmosphere. Lead acetate trihydrate, zirconium acetate pentahydrate, titanium isopropanoxide, acetic acid, and 2-methoxyethanol were used as the source materials and solvents. The concentration of the final solution of PbZr$_{0.52}$Ti$_{0.48}$O$_3$ was adjusted to 0.2 mol/L by adding solvents. Similarly, 0.05 mol/L PbTiO$_3$, PbZr$_{0.25}$Ti$_{0.75}$O$_3$, PbZr$_{0.52}$Ti$_{0.48}$O$_3$, PbZr$_{0.75}$Ti$_{0.25}$O$_3$ solutions were prepared. 10% extra lead was added to compensate for the lead loss during the annealing of the film (for all the sols). The films of PZT were coated on Pt/Ti/SiO$_2$/Si substrates directly, or on Pt(111)/Ti/SiO$_2$/Si substrates containing the seed islands of PbTiO$_3$, PbZr$_{0.25}$Ti$_{0.75}$O$_3$, PbZr$_{0.52}$Ti$_{0.48}$O$_3$, PbZr$_{0.75}$Ti$_{0.25}$O$_3$ annealed at 600 °C, respectively. After spinning coating, the coated films were dried at 180 °C for 3 min and pyrolyzed at 400 °C for 5 min. These processes were repeated four times to obtain desired film thickness. After that, the films were finally annealed at 600 °C for 5 min using RTP.

**Characterization**

To investigate the electric properties of the films, Pt top electrodes with a diameter of 200 μm were deposited on the films by DC (direct current) sputtering (ETD-3000, China). The microstructure characterization of the films was represented by X-ray diffractometer (XRD, Rigaku D/MAX-2500PC, Japan) with Cu Kα radiation. The dielectric properties of the films were measured using semiconductor characterization system (Agilent B1500A, USA). The mesoscopic domain switching capability of the films was represented by PFM (Bruker Multimode). The ferroelectric properties were analyzed by ferroelectric test systems (Radiant Technologies Precisions workstations RT 66A, USA). Among them, all P–E hysteresis loops were measured by two tips at a frequency of 1 kHz, and the polarization fatigues were determined by a pulse signal of amplitude 7.5 V, 100 kHz.

**Results and discussion**

To characterize the thickness of the films, we performed cross-sectional SEM tests on all samples. As
shown in supplementary material Fig. S1, the thickness of all films is around 150 nm. All the Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin films with Pb(Zr$_x$Ti$_{1-x}$)O$_3$ seed islands show pure perovskite phase with no other impurity phases [23], as demonstrated by the XRD patterns shown in Fig. 1b. From these X-ray diffraction patterns, we learn the pure Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin film and the Pb(Zr$_x$Ti$_{1-x}$)O$_3$ thin films with Pb(Zr$_x$Ti$_{1-x}$)O$_3$ seed islands except for PTO prefer to performing textured (111)-orientation along out of the plane with weak (110) peak. This is because, on the one hand, with the increase in Zr content, PZT thin films tend to form (111)-oriented rhombohedral structure, on the other hand, its orientation may be controlled by the orientation of the Pt bottom electrode, (111)-oriented Pt bottom electrodes are easier to obtain (111) preferred orientation PZT thin film [24]. The Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin film with the PTO seed islands is mainly textured along the (100) direction with a weak (110) peak and the crystal structure tends to be tetragonal phase since the tetragonal PTO seed islands with (100)-orientation have the minimum electrostatic energy due to the in-plane polarization. In addition, an extra peak can be observed at around 36° for all thin films, which may be due to the instrument. All PZT films grow in the (111) directional texture to ensure that the films own the expected crystal structure [25].

The polarization–electric field hysteresis loops (P–E) and the capacitance–electric field curves (C–E) on all the Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin films with Pb(Zr$_x$Ti$_{1-x}$)O$_3$ seed islands were measured at a frequency of 1 kHz and 1 MHz at room temperature, which are shown in Fig. 2a and c, respectively. All films exhibit ferroelectric switching loop behavior and representative “butterfly” curves (C–E) which indicate excellent ferroelectric properties. Obviously, the P–E loops and the C–E curves for these PZT thin films modified by seed islands are better than the ones without seed islands. The maximum polarization, remnant polarization, and coercive field of the films are summarized in Fig. 2b, the $2P_r$ of these films modified by the PZT seed islands with different ratios of Zr/Ti (0/100, 25/75, 52/48, 75/25) are 55.9 μC/cm$^2$, 53 μC/cm$^2$, 56.5 μC/cm$^2$ and 49.5 μC/cm$^2$, respectively, and the remnant polarization is enhanced a lot compared with pure PZT thin film (42 μC/cm$^2$). When the Zr/Ti ratio of the seed islands is 52/48, the regulation of the seed islands on PZT ferroelectric thin films reaches the maximum, while the coercive field is the smallest ($2E_c = 138$ kV/cm), the remnant polarization and the maximum polarization reaches the maximum($2P_r = 56.5$ μC/cm$^2$, $2P_{\text{max}} = 161.4$ μC/cm$^2$). The remnant polarization of the Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin film with Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ seed islands is enhanced by 34.5% compared with pure PZT thin film. Thus, the seed islands with the same ratio of PZT sol–gel can remarkably enhance the ferroelectric properties of the PZT films. This is because the addition of the seed islands improves interfacial crystallization and optimizes the thin film microstructure, which is crucial to getting access to high performance [26]. In addition, we also found that the coercive field of the PZT thin film with the Zr/Ti (52/48) seed islands in the (111) texture direction was significantly smaller than that of the PZT thin film with the PTO seed islands in the (100) texture direction. This is because when an adequate electrical field perpendicular to the surface of the thin film is applied on, the domain of the (111)-oriented
PZT thin film is parallel to the direction of the electric field, while the original polarization along the a-axis of the (100)-oriented PZT thin film needs to rotate to the c-axis direction under the action of the electric field, that is, from the $P_c$ polarization direction to the $P_a$ polarization direction, as shown in Fig. 3. The rotation of the polarization is associated with the lattice distortion which gives rise to remarkable local stress in the thin films [27]. Furthermore, the rotation displacement of the polarization of the (100)-textured PZT films is greater than that of the (111)-textured PZT thin films, which makes the domain switching more difficult and requires a larger coercive electric field.

Furthermore, the polarization fatigues of all the Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin films with Pb(Zr$_x$Ti$_{1-x}$)O$_3$ seed islands are displayed in Fig. 2d. It can be seen that the seed islands have a great influence on the fatigue resistance of the films. When the ratio of the seed islands is 52/48, the thin film shows the most superior fatigue resistance can reach $10^9$ cycles without attenuation, which is attributed to the high quality and stability of the film structure. In addition, the $P_r/P_1$ value of PZT(52/48)/PZT(52/48) thin film reflects an increasing trend in the $10^7$–$10^9$ fatigue cycle interval. According to the change of fatigue cycle test curves of PTO/PZT(52/48) thin film and PZT(75/25)/PZT(52/48) thin film, we believe that the increased $P_r/P_1$ value of PZT(52/48)/PZT(52/48) thin film may be caused by the increase in leakage current in the ferroelectric capacitor before breakdown. The activation energy for the nucleation of the perovskite phase of PZT thin film, $E_A(N)$, is 441 kJ/mole and that for the crystal growth, $E_A(CG)$, is 112 kJ/mole. Therefore, the high value of activation energy for nucleation is the primary barrier for
the crystallization of perovskite phase PZT. PZT seed islands were crystallized into the perovskite phase before the deposition of PZT thin film and provided sufficient nucleation sites for the PZT thin film [28]. Therefore, it was considered that PZT seed islands could enhance the transformation of the perovskite phase in PZT thin film without the need for $E_A(N)$ to a great extent, thus promoting the growth of subsequent film, shown in Fig. 1a. What’s more, the reason why the best ferroelectric and dielectric properties appear in the ones whose ratio of Zr/Ti get to 52/48 is due to this ratio of the seed islands is the same with the following covered PZT (Zr/Ti = 52/48) thin film, as a result, it can induce the directional crystallization of PZT film to the greatest extent, and greatly reduce the defects, strain, and dead layer caused by interface mismatch, making the interface between seed islands and the thin film better than others [29, 30].

In order to further investigate the quality of PZT thin films, dielectric properties testing and leakage current density testing are essential. Figure 4a shows a dielectric spectrum ($\varepsilon$–$F$) of PZT thin films with seed islands in different Zr/Ti ratios. As it can be seen from the figure, they were measured at room temperature with a frequency range from 1 kHz to 1 MHz function varies. The dielectric constants of all the Pb(Zr$_{0.52}$Ti$_{0.48}$)O$_3$ thin films slowly decrease linearly as the logarithm of the frequency increases, which indicates that the dielectric constant of the films is mainly controlled by the domain wall pinning [31]. The dielectric constants of films modified by seed islands are all higher than no modified one [32]. When the Zr/Ti ratio of seed islands is 52/48, the dielectric constant reaches the maximum, and the dielectric constant is 960 at the frequency of 1 kHz, which is about twice as high as that of pure PZT film. Adding seed islands can reduce the overall activation energy of the PbZr$_{0.52}$Ti$_{0.48}$O$_3$ thin films for crystallization, and the interfacial crystallization is improved. At the same time, the existence of seed islands can effectively reduce the diffusion between the film and the substrate, which decreases the dead layer and makes the grain size of the film more uniform with fewer defects [33]. The external contribution of the dielectric constant of the PZT thin film is mainly attributed to the domain wall motion. The improvement of the crystallinity of the PZT thin film with the seed islands and the reduction of its internal defects decrease the pinning effect on the crystal grains in the relative volume caused by the defects, which makes the movement of the domain wall easier, and thus the dielectric constant increase [34-36]. Figure 4b shows the leakage current densities as a function of the electric field with the Pt top electrodes for the above PZT thin films. It is obvious that the leakage current densities of PZT thin films modified by seed islands are smaller than the one without seed islands. Furthermore, we also measured

![Figure 3](image-url)  
**Figure 3** The domain polarization direction of a (111)-oriented and b (100)-oriented PZT thin films.
the leakage current performance of the films at higher electric field strength. As indicated by Fig. S2 in supplementary materials, similar to the leakage current density–electric field curves in Fig. 4b, the leakage current densities of the PZT films modified by Pb(Zr\(_{x}\)Ti\(_{1-x}\))O\(_3\) seed islands at high electric field strength are also lower than that of the pure PZT film. This is because the addition of the extremely low concentration of PZT(Zr/Ti = 52/48) as the seed islands provides nucleation points for subsequent crystallization of PZT films, which reduces the activation energy of film nucleation and growth. Besides, it can also maintain high surface energy, which is conducive to the diffusion of thin films surface at low temperature and allows the films and oxygen to be in full contact. Thus, the crystallization transition speed of the thin film is increased, the oxygen vacancy concentration in the thin film is decreased, which increases the density of the thin film and reduces the defects in the thin film. And finally, the leakage current density of the thin film is reduced, which corresponds to the experimental results of fatigue resistance in Fig. 2d [37, 38].

In order to characterize the mesoscopic domain switching of the thin films, piezoresponse force microscopy (PFM) is an ideal tool for both probing and switching the local ferroelectric polarization at the nanoscale. The box-in-box switched patterns were written on the five samples with the application of the electric field via a conducting tip. A tip bias of \(-15\) V was applied to pole the 12 \(\mu\)m x 12 \(\mu\)m square region followed by another poling with a tip bias of +15 V in the central area of 6 \(\mu\)m x 6 \(\mu\)m. The out-of-plane polarization signals of these five samples are shown in Fig. 5a–e, which show clear bright and dark contrast regions. As can be seen from the figures, the switching contrasts of the films with the seed islands are larger than that of the film without the seed islands. In particular, the PZT thin films with Zr/Ti ratio of 52/48 of the seed islands can be switched when the tip voltage is 12 V. This result suggests that adding PZT seeds into the PZT thin films can promote the domain switching. This is because the addition of PZT seed islands regulates the interface of PZT film, which reduces the internal defects of the film, and the pinning effect on the domain wall is weakened due to fewer defects, so the domain is easier to be switched [39, 40]. This result can also be reflected in Figs. 2a and 4b. Figure 5f shows representative local PFM phase hysteresis loops. The square loops demonstrating a 180° change in the PFM phase confirm the excellent ferroelectric switching nature of these five samples. The 2\(V_c\) values and phase contrasts of the PZT films corresponding to Fig. 5f are summarized in Table 1. From the PFM loops, the 2\(V_c\) of the one modified by PZT (Zr/Ti = 52/48) is the lowest, which also confirms the film modified by PZT (Zr/Ti = 52/48) can accelerate the domain switch [41, 42] and enhance the ferroelectric properties, which is consistent with the previous macro results.

Conclusions

In conclusion, PZT thin films modified by PZT seed islands with different Zr/Ti ratios were successfully fabricated on Pt(111)/Ti/SiO\(_2\)/Si substrates using the
sol–gel process. Compared with pure PZT films, the electrical properties of PZT films modified by PZT seed islands with different Zr/Ti ratios, including ferroelectric properties and dielectric properties, have been improved to a large extent. Especially when the ratio of Zr/Ti is 52/48 for the PZT seed islands, the leakage current density attains the minimum, and ferroelectric and dielectric properties are the best. Compared with other materials as the seed islands, using the same material as the seed islands can form a better interface. This work reports a new method to improve the electrical properties of ferroelectric thin films, which will promote the development of new high-performance ferroelectric memory.

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**Figure 5** PFM phase diagrams for PZT thin films

- **a** with no seed islands,
- **b** with seed islands of PTO,
- **c** with seed islands of Zr/Ti ratio of 25/75,
- **d** with seed islands of Zr/Ti ratio of 52/48,
- **e** with seed islands of Zr/Ti ratio of 75/25, and
- **f** phase–voltage curves of PZT thin films.

**Table 1** The $2V_c$ and phase contrast of several films corresponding to Fig. 5f

| PZT thin films       | PZT(52/48) | PTO/PZT(52/48) | PZT(25/75)/PZT(52/48) | PZT(52/48)/PZT(52/48) | PZT(75/25)/PZT(52/48) |
|----------------------|------------|----------------|----------------------|----------------------|----------------------|
| $2V_c$ (V)           | 7.0        | 10.2           | 9.8                  | 6.8                  | 10.0                 |
| Phase contrast (deg) | 166.5      | 173.6          | 165.4                | 173.4                | 165.4                |
Declarations

Conflict of interest  The authors declare no competing financial interest.

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