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Effects of interface number on the temperature and frequency dependence of the properties of $Pb(Zr_{0.52}Ti_{0.48})O_3/Ba(Mg_{1/3}Ta_{2/3})O_3$ thin films

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ABSTRACT

In this paper, $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) and $Ba(Mg_{1/3}Ta_{2/3})O_3$ (BMT) thin films were prepared by sol-gel method and aqueous solution-gel method, respectively. PZT/BMT thin films with different interface numbers were prepared and the effects of interface number on the temperature and frequency dependence of PZT/BMT thin films were investigated. The interface number can improve the temperature stability of PZT/BMT thin films in the 85 to 205 °C range. As the temperature increases, there is an increase in the remanent polarization for the PZT/BMT thin films and the increasing rate of the remanent polarization decreases with the increase of interface number. As the frequency increases, the remanent polarization for the PZT/BMT thin films decreases and the decline rate of remanent polarization slows with the interface number increasing, indicating that interface number can improve the frequency stability of PZT/BMT thin films.

1. Introduction

In recent years, more and more attention has been attracted to the ferroelectric thin films due to their wide range applications [1,2]. These applications may be used over a wide temperature and frequency ranges but unfortunately ferroelectric thin films exhibit poor temperature and frequency stability. Hence, it is significantly important to improve the temperature and frequency stability of ferroelectric thin films. Many studies have shown that it is an effective method to add some layers with good temperature and frequency stability to improve the stability of thin films [3,4]. Lee et al. [5] have found that the temperature stability of $MgTiO_3/CaTiO_3$(MT/CT)thin films was related to the thickness of CT thin film and the temperature coefficient of dielectric constant at microwave frequency was changed from positive to negative values by controlling the thickness of CT thin film. Most studies were focused on the effects of the thickness on the temperature dependence of multilayer thin films. The effects of interface number on the temperature and frequency dependence of multilayer thin films were rarely investigated. In fact, the interface number has an important influence on the properties of the multilayer thin films [6–10], but few investigations studied the relationship between the interface number and temperature or frequency stability of the multilayer thin films. Therefore, it is necessary to study the effects of interface number on the temperature and frequency dependence of the multilayer thin films.

In this paper, $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) and $Ba(Mg_{1/3}Ta_{2/3})O_3$ (BMT) thin films were prepared by sol-gel method and aqueous solution-gel method, respectively. PZT/BMT thin films with different interface numbers were prepared and the effects of interface number on the temperature and frequency dependence of PZT/BMT thin films were investigated.

2. Experimental details

PZT thin films were prepared by sol-gel method and BMT thin films were prepared by aqueous solution-gel method according to the literatures [11,12]. The concentration of the PZT and BMT solution was adjusted to 0.4 M, 0.1 M, respectively. In order to compensate for lead loss during heat treatment, a 15 mol% excess of lead acetate was added to the PZT solution.

The PZT/BMT thin films with different interface numbers were deposited as shown in Fig. 1. Two series of samples have the same thickness, denoted as PPBB, PBPB.

The PZT/BMT thin films were deposited on $Pt/Ti/SiO_2/Si(100)$ substrates by a spin coating method at 4000 rpm for 30 s. Each BMT layer was dried at 180 °C and 380 °C for 2 min, respectively. And then a pre-annealing process was conducted at 600 °C for 10 min. Each PZT layer was dried at 150 °C and 350 °C, respectively, and then annealed at 600 °C for 5 min. Finally, the PZT/BMT thin films were annealed at

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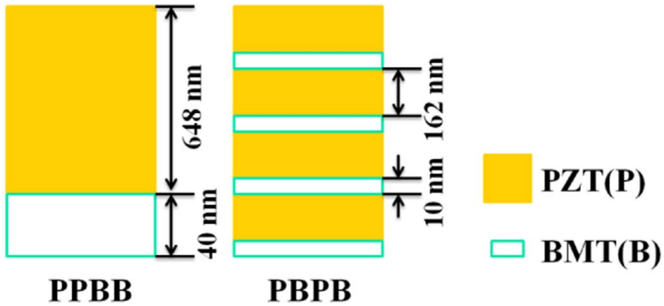

Fig. 1. Schematic structure of PZT/BMT thin films with different interface numbers.

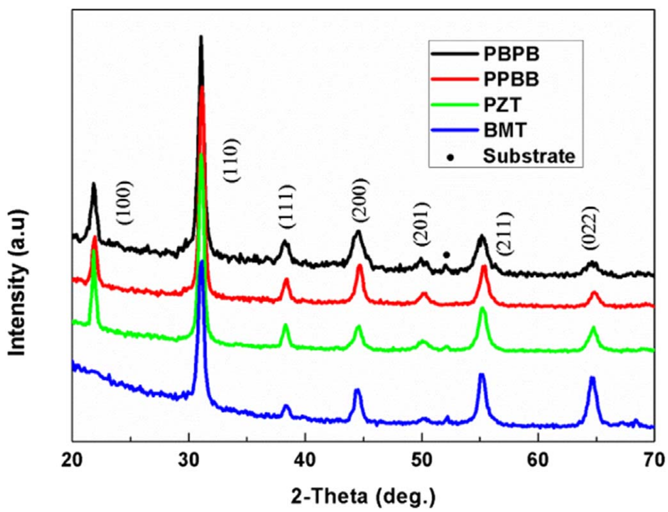

Fig. 2. XRD patterns of PZT thin films, BMT thin films and PZT/BMT thin films with different interface numbers.

700 °C for 60 min.

The crystalline phase of PZT/BMT thin films was studied by X-ray diffraction (XRD, PANalytical X'Pert PRO) by using CuKα (λ = 1.5406 Å) radiation. The scanning range was 10–70° with a step size of 0.02° and a measuring time of 1 s. The cross-sectional and surface micrographs of PZT/BMT thin films were characterized by field emission-scanning electron microscopy (FE-SEM, ULTRA Plus-43-13) and the operating voltage is 5.0 kV. The dielectric properties of PZT/BMT thin films were measured at different temperatures by precision impedance analyzer (Agilent 4294A, Agilent technologies, USA). The ferroelectric properties and leakage current-voltage (I–V) characteristics of PZT/BMT thin films were carried out with precision workstation (Radiant Technologies, USA) at different temperatures and frequencies. The applied field was a triangle signal and the samples experienced two periods of the field during ferroelectric hysteresis loops (P-E loops) measurement at different temperatures and frequencies.

3. Results and discussion

3.1. Microstructures and morphology

Fig. 2 shows the XRD patterns of PZT thin films, BMT thin films and PZT/BMT thin films. The XRD shows that all the thin films are polycrystalline with a perovskite structure. It can be seen that all the thin films are fully crystallized without any second phase. PZT and BMT thin films have some similar main peaks which are very close. Meanwhile, compared with BMT thin films, PZT thin films have a characteristic peak, which is (100) peak. Therefore, apart from the characteristic peak, it is difficult to distinguish the main peaks of PZT and BMT thin films.

Fig. 3 shows the cross-sectional FE-SEM images of PZT/BMT thin films with different interface numbers. All the PZT/BMT thin films have a dense microstructure without any crack across the film thickness. In Fig. 3(a), the total thicknesses of PZT and BMT thin films are about 648 nm, 40 nm, respectively. In Fig. 3(b), each PZT layer is about 162 nm. However, each BMT layer is too thin to be directly observed. The thicknesses of PZT and BMT thin films were controlled by adjusting the number of coatings. The average thickness of each BMT layer is about 10 nm.

3.2. Temperature stability

Fig. 4 shows the temperature dependence of the dielectric properties of PZT/BMT thin films with different interface numbers at the frequency of 1 MHz and the test temperature ranges from 25 to 205 °C. The dielectric constant of PBPB thin films at 25 °C is much higher than that of PPBB thin films and the dielectric loss of PBPB thin films is much lower than that of PPBB thin films, indicating that the increase of interface number can enhance the dielectric properties of PZT/BMT thin films. According to the reference [13], the dielectric constant of dielectric/dielectric thin films increases with the increase of interface number, which is also true to the ferroelectric/dielectric thin films. Both the dielectric constant and dielectric loss of PPBB and PBPB thin films increase with the temperature increasing and the increasing rate of the dielectric constant and dielectric loss of the PPBB thin films is much faster than that of PBPB thin films in the 85 to 205 °C range, indicating that the interface number is advantageous to the temperature stability of PZT/BMT thin films.

Fig.5 shows the leakage current-voltage (I–V) characteristics of PZT/BMT thin films with different interfaces. The leakage currents of all the thin films are less than 2×10^{-8} A at the maximum applied voltage. It can be seen that all the thin films exhibit excellent leakage current properties. The leakage currents of PBPB thin films are lower than that of PPBB thin films.

Figs. 6 and 7 show the ferroelectric properties of PZT/BMT thin films with different interface numbers at the frequency of 10 Hz and the test temperature ranges from 25 to 145 °C. The remnant polarization

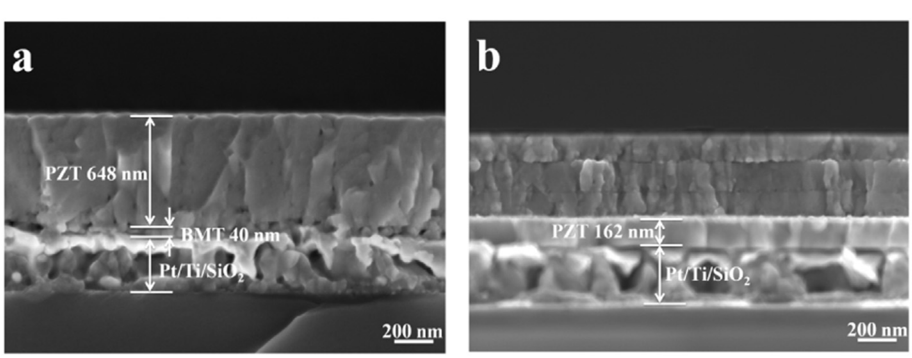

Fig. 3. Cross-sectional FE-SEM images of PZT/BMT thin films: (a) PPBB thin films, (b) PBPB thin films.