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Effect of the PbO_x thickness on the microstructure and electrical properties of PLT thin films prepared by RF magnetron sputtering

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Abstract

The $(Pb_{0.90}La_{0.10})Ti_{0.975}O_3$ (PLT) thin films with different thicknesses of PbO_x buffer layers were deposited on the $Pt(111)/Ti/SiO_2/Si(100)$ substrates by RF magnetron sputtering technique. The PbO_x buffer layer leads to the (100) orientation of the PLT thin films. Effects of the PbO_x thickness on the microstructure and electrical properties of the PLT thin films were investigated. The experimental results show that the PbO_x thickness plays an important role on the orientation, phase purity, domain structure, and electrical properties of the PLT thin films. The PLT thin films with proper PbO_x thickness possess highly (100) orientation, high phase purity, strong intensity of out of plane polarization, and good electrical properties. It is concluded that the PbO_x thickness between PLT thin films and Pt coated Si substrate is very critical to obtain good electrical properties.

Keywords: Ferroelectric thin films; RF magnetron sputtering; Buffer layer; Microstructure; Electrical properties

1. Introduction

Ferroelectric thin films have attracted considerable attention in recent years because of their practical or potential applications in various fields, such as ferroelectric random access memories, pyroelectric sensor, and so on [1,2]. Among the researches, lanthanum doped lead titanate thin films possess good electrical properties [3], which is one of the most extensively studied materials [4–6].

Many factors may affect electrical properties of ferroelectric films, such as the orientation, composition, buffer layers, and so on [7–14]. Among these, the buffer layer is an important factor for improving electrical properties of the films [11–14]. Moreover, it was also reported that the thickness of buffer layer is critical to obtain good electrical properties of the films [10]. As a result, it is necessary to systemically study the effect of the thickness of buffer layers on the microstructure and electrical properties of the films.

In this work, effects of the thickness of PbO_x buffer layer on the orientation, phase purity, domain structure, and electrical properties of $(Pb_{0.90}La_{0.10})Ti_{0.975}O_3$ (PLT) thin films, which were *in situ* deposited on the $Pt(111)/Ti/SiO_2/Si(100)$ substrates by RF magnetron sputtering technique, were investigated.

2. Experimental

The PbO_x buffer layers with different thicknesses were firstly *in situ* deposited on the $Pt(111)/Ti/SiO_2/Si(100)$ substrates at the substrate temperature of 650 °C, and then PLT thin films were *in situ* prepared on the $PbO_x/Pt(111)/Ti/SiO_2/Si(100)$ substrates by RF magnetron sputtering at the substrate temperature of 600 °C. The targets of PLT and PbO were prepared by conventional ceramic method. The sputtering power was fixed at 50 W for PbO and 80 W for PLT. The distance between the target and substrate is 5 cm. The deposition atmosphere for the PbO_x buffer layers and PLT thin films is with the ratio of oxygen (O): argon (Ar) of 10 sccm: 40 sccm under a working pressure of 2.0 Pa. The thickness of PbO_x buffer layer was estimated by the deposition time according to the average deposition rate for the layer.

X-ray diffraction (XRD) characterization of the films was performed by using Cu Kα radiation ($\lambda = 1.54178$ Å) in the θ–2θ scan mode (DX-1000, Dandong, China). Surface morphologies

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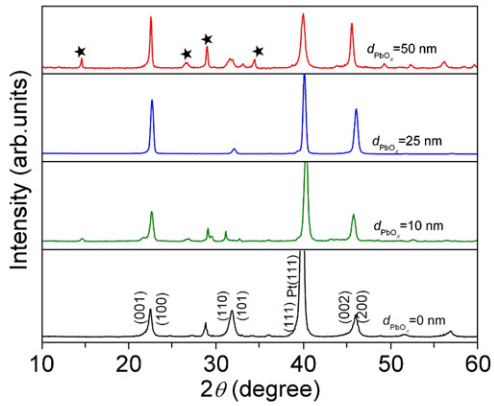

Fig. 1. XRD patterns of the PLT thin films with different PbO_x thicknesses (★: pyrochlore phase).

and domain structures of the films were measured using a modified scanning probe microscope (Seiko Instruments, SPA-300HV, Japan), which operated in the piezoresponse force microscopy mode (PFM). PFM allows sample topography and out-of-plane polarization (OPP) images to simultaneously be recorded. Au coated Si_3N_4 cantilever (Seiko) with an elastic constant of 1.4 N/m and an integrated tip of about 20 nm diameter. During the scanning, the same ac voltage with the amplitude of 5.0 V and the frequency of 9 kHz was applied on the films. In order to measure the electrical properties of the films, dot-type gold electrodes with an area of 20×10^{-4} cm^2 were deposited by dc sputtering, which form Au/PLT/PbO_x/Pt(111)/Ti/SiO_2/Si(100) stacked capacitors. The dielectric properties of the capacitors were measured by HP4194A impedance analyzer. The hysteresis loops of polarization (P) as a function of applied electric field (E) (P–E curve) were evaluated using Radiant Precision Ferroelectric Measurement System (RT2000 Tester, USA). The system for measuring the pyroelectric coefficients (p) of ferroelectric thin films was developed using the dynamic method by the Shanghai Institute of Technical Physics, Chinese Academy of Sciences [15]. All the measurements of dielectric, ferroelectric, and pyroelectric properties of the films were carried out at the Institute.

3. Results and discussion

Fig. 1 shows the XRD patterns of the PLT thin films with different PbO_x thicknesses. All PLT thin films with a PbO_x buffer layer possess (100) orientation, but have different degree of preferential (100) orientation. The preferential (100) orientation parameter, α_{hkl}, can be calculated by the following formula:

$$\alpha_{hkl} = I_{hkl}/\Sigma I_{hkl} \quad (1)$$

where I_{hkl} is the relative intensity of the corresponding diffraction peaks. The calculated preferential (100) orientation of the PLT thin films with about 25 nm-thickness PbO_x buffer layer is highest (α_{hkl}=93%). As a result, the (100)-oriented PbO_x buffer layer leads to the (100) orientation of the PLT thin films, which had been described elsewhere [12]. For the lead-based films with PbO_x buffer layer *in situ* deposited, the nuclei

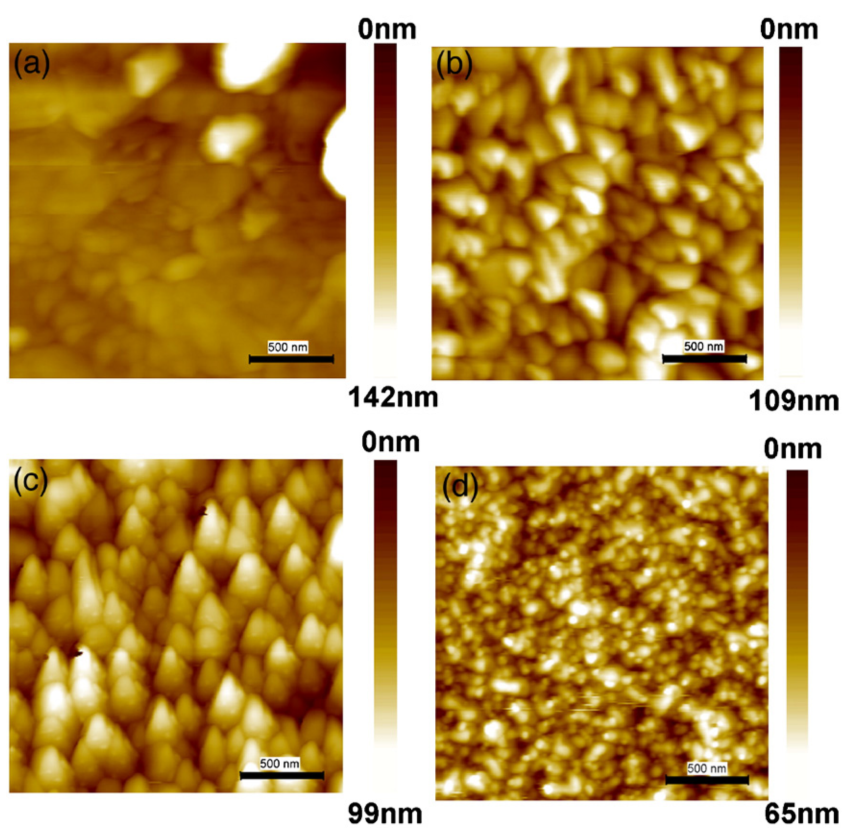

Fig. 2. AFM images (2 μm × 2 μm) of the PLT thin films with (a) 0 nm; (b) 10 nm; (c) 25 nm; (d) 50 nm-thickness PbO_x buffer layer.