

Enhanced ferroelectric properties of $\text{Pb}(\text{Zr}_{0.80}\text{Ti}_{0.20})\text{O}_3/\text{PbO}$ thin films prepared by radio frequency magnetron sputtering

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Abstract

The $\text{Pb}(\text{Zr}_{0.80}\text{Ti}_{0.20})\text{O}_3$ (PZT) thin films with and without a PbO buffer layer were deposited on the $\text{Pt}(1\ 1\ 1)/\text{Ti}/\text{SiO}_2/\text{Si}(1\ 0\ 0)$ substrates by radio frequency (rf) magnetron sputtering method. The PbO buffer layer improves the microstructure and electrical properties of the PZT thin films. High phase purity and good microstructure of the PZT thin films with a PbO buffer layer were obtained. The effect of the PbO buffer layer on the ferroelectric properties of the PZT thin films was also investigated. The PZT thin films with a PbO buffer layer possess better ferroelectric properties with higher remnant polarization ($P_r = 25.6\ \mu\text{C}/\text{cm}^2$), and lower coercive field ($E_c = 60.5\ \text{kV}/\text{cm}$) than that of the films without a PbO buffer layer ($P_r = 9.4\ \mu\text{C}/\text{cm}^2$, $E_c = 101.3\ \text{kV}/\text{cm}$). Enhanced ferroelectric properties of the PZT thin films with a PbO buffer layer is attributed to high phase purity and good microstructure. © 2007 Elsevier Ltd. All rights reserved.

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1. Introduction

Ferroelectric thin films have attracted much attention in recent years, because of their practical or potential applications in various fields, especially for ferroelectric random access memories (FeRAM) [1,2]. Among the researchers, lead zirconate titanate [$\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT)] thin films have been one of the most extensively studied materials for nonvolatile random access memory application due to large remnant polarization and low coercive field [3,4].

Recently, a few papers [5,6] reported that the rhombohedral ($x < 0.48$) PZT films possessed several important ferroelectric properties including low coercive field, large remnant polarization, low leakage current density, which are correlative to FeRAM capacitors, moreover, are superior as compared with those in the MPB region or in the tetragonal side of the MPB. The phase transformation temperature of PZT thin films usually depends on the Zr/Ti ratio, so the PZT films with higher Zr/Ti ratio require higher transformation temperature [7,8]. However, high temperature processing is easy to cause the diffusion of Pb from the films to the substrate, and results in undesirable pyrochlore phase. As a result, it is very important for fabricating Zr-rich PZT thin films to control the perovskite phase purity.

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In this work, the $\text{Pb}(\text{Zr}_{0.80}\text{Ti}_{0.20})\text{O}_3$ (PZT) thin films with and without a PbO buffer layer were deposited on the $\text{Pt}(1\ 1\ 1)/\text{Ti}/\text{SiO}_2/\text{Si}(1\ 1\ 1)$ substrate by radio frequency (rf) magnetron sputtering method. The effect of PbO buffer layer on the microstructure and ferroelectric properties of the PZT thin films is mainly investigated.

2. Experimental

In this work, we report the preparation of $\text{Pb}(\text{Zr}_{0.80}\text{Ti}_{0.20})\text{O}_3$ thin films with and without a PbO buffer layer on the $\text{Pt}(1\ 1\ 1)/\text{Ti}/\text{SiO}_2/\text{Si}(1\ 0\ 0)$ substrate by rf magnetron sputtering at the substrate temperature of $450\ ^\circ\text{C}$, and followed by annealing in air by conventional furnace annealing (CFA). Sputtering targets of PbO and PZT thin films were made by a conventional mixing-oxides method, 10 mol% excess PbO was added to PZT target for compensating for the loss of Pb in PZT targets. Sputtering of PbO buffer layer and PZT thin films was carried out at a power of 50 and 80 W, respectively. A high purity sputtering gas is composed of 20% O_2 + 80% Ar under a working pressure of 2.0 Pa. The PZT thin films without a PbO buffer layer were sputtered with the PZT targets under the same conditions as the PZT films with a PbO buffer layer for comparison.

X-ray diffraction (XRD) characterization of the films was performed by using Cu $\text{K}\alpha$ radiation ($\lambda = 1.54178\ \text{\AA}$) in the mode of θ – 2θ scan (DX-1000, Dandong, China). The cross-sectional images of the films were observed by scanning electron microscopy (SEM, JSM-5900, Japan). The surface morphology was studied by atomic force microscope (AFM, SPA-400, Japan). The chemical composition analysis of the films was carried out by energy dispersive X-ray (EDX) in field emission scanning electron microscope (FE-SEM, HITACHI S4800, Japan). In order to measure the ferroelectric properties of the films, dot-type gold electrodes with an area of $20 \times 10^{-4}\ \text{cm}^2$ were deposited by dc sputtering, which form stacked capacitors. The hysteresis loops were evaluated by using Radiant Precision Ferroelectric Measurement System (Precision Workstation Tester, USA).

3. Results and discussion

The preparation of Zr-rich PZT thin films on Pt-coated Si substrate is not easily straight to form perovskite phase, but the pyrochlore phase always forms preferentially during heat treatment of film [9]. In order to improve the phase purity of PZT thin films, in this work, the PbO was used for the buffer layer, the PZT thin films with a PbO buffer layer were deposited at the substrate temperature of $450\ ^\circ\text{C}$, and followed by annealing at $650\ ^\circ\text{C}$ in air by conventional furnace annealing. Fig. 1(a and b) shows the XRD patterns of the PZT thin films with and without a PbO buffer layer. It is found that the PZT thin films with a PbO buffer layer possess pure perovskite structure with random orientation. However, the PZT thin films without a PbO buffer layer possess a mixture of pyrochlore and perovskite structure. Such a phenomenon is because the lead element of the PbO buffer layer is also one of the main elements of PZT thin films, and the lead element of the films may be partially volatile during film's deposition. We regard in this case that the lead

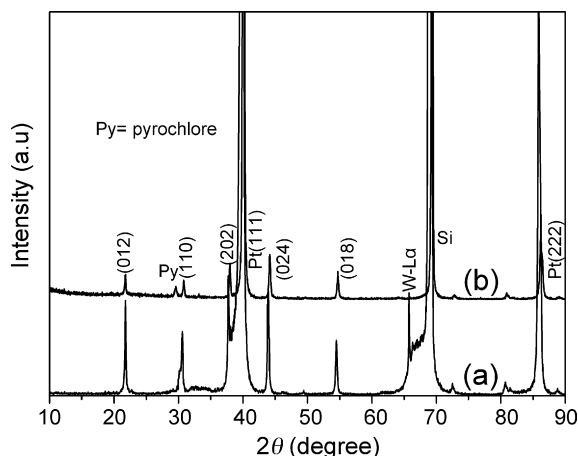


Fig. 1. X-ray diffraction patterns of the PZT thin films with (a) and without (b) a PbO buffer layer.

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