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The effects of deposition temperature on structure and dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$ thin films produced by pulsed laser deposition

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Abstract

The effects of deposition temperature on orientation, surface morphology and dielectric properties of the thin films for $Ba_{0.6}Sr_{0.4}TiO_3$ thin films deposited on Pt/Ti/SiO_2/Si substrates by pulsed laser deposition were investigated. X-ray diffraction patterns revealed a (2 1 0) preferred orientation for all the films. With rising substrate temperature from 650 °C to 700 °C, the crystallinity and crystal grain size of the films increase, the relative dielectric constant increases, but the dielectric losses have not obvious difference. The film deposited at 350 °C and annealed at 700 °C has strongly improved roughness and dielectric permittivity compared with the film only deposited directly at 700 °C. Three distinct relaxation processes within tan(δ) were found for the $Ba_xSr_{1-x}TiO_3$ film: a broadened process of the film relaxation, an intermediate peak which originates from Maxwell–Wagner–Sillars polarization, and an extremely slow process ascribed to leak current. The complex dielectric permittivity and loss can be fitted by an improved Cole–Cole model corresponding to a stretched relaxation function.

Keywords: Pulsed laser deposition; BST thin film; Oxygen pressure; Dielectric relaxation

1. Introduction

$Ba_xSr_{1-x}TiO_3$ (BST) thin film is a continuous solid solution between $BaTiO_3$ and $SrTiO_3$ over the whole composition range. The solid solution of BST is known to show a decrease in transition temperature with increasing strontium content. In ceramics the rate of decrease of transition temperature with strontium content has been reported as 3.75 °C/mol% of $SrTiO_3$ [1]. In pulsed laser ablated films Gim et al. has reported this rate as 4.59 °C/mol% of $SrTiO_3$ [2]. Because of their potential applications, BST thin films have been attracting a great deal of attention in decoupling capacitors, storage capacitors, giga-bit dynamic random access memories and tunable microwave devices. The large electric field-dependent dielectric constant can be used for devices such as tunable oscillators, filters and phase shifters [3–6]. In such devices, it is desirable to have a high dielectric tunability in a certain electric field range and low dielectric loss. The dielectric constant and tunability of capacitance are sensitive to the relative crystal orientation of the films [7]. There are various methods for changing the orientation of the ferroelectric films, such as the control of heat treatment conditions and the deposition ambience [7–10].

In this work, $Ba_{0.6}Sr_{0.4}TiO_3$ thin films were fabricated by pulsed laser deposition (PLD) on Pt/Ti/SiO_2/Si substrates at different growth temperatures. The effects of growth temperature on structure, surface morphology and dielectric properties of PLD-derived thin films are presented and discussed.

2. Experimental details

The $Ba_{0.6}Sr_{0.4}TiO_3$ thin films were grown on Pt/Ti/SiO_2/Si substrates produced by pulsed laser deposition using the KrF (248 nm wavelength) excimer laser system (Lambda Physik LPX 300 cc) with a repetition rate of 4 Hz and 25 ns in pulse duration. The laser beam was focused by a quartz lens to a fluency of approximately 2.5 J/cm^2 and directed at an angle of 45° on the target. The target, a stoichiometric BST ceramic disc of 25 mm diameter × 5 mm thick, was rotated during the ablation process to reduce non-uniform erosion. The deposition rate was 20 nm/min. The substrate of 5 mm × 15 mm was placed parallel to the target at a distance of 40 mm.

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At first, in order to study the deposition temperature effects, three films with the thickness of 300 nm were deposited in oxygen pressure of 0.01 Pa and at substrate temperature of 650 °C, 680 °C and 700 °C. At last, for the study of annealing effects, films with the same thickness were deposited in oxygen pressure of 0.01 Pa (1) at substrate temperature of 700 °C (BST700) directly and (2) at substrate temperature of 350 °C and annealed at 700 °C for 30 min (BST350).

The structure of the BST thin films were analyzed by X-ray diffractometer (XRD, d/max ultima iii, 1.5406 nm) using a characteristic X-ray of Cu Kα. The microstructure and surface roughness characterizations were performed by atomic force microscopy (AFM) (type: HL-II, China) using a contact mode. Pt electrodes (1 mm in diameter) were deposited on top of the BST films by dc sputtering through a metallic mask. Dielectric complex permittivity of measurements for the Pt/BST/Pt system was performed using the HP 4192A LF impedance analyzer.

3. Results and discussion

The XRD patterns of the BST thin films deposited in oxygen pressure of 0.01 Pa and at different substrate temperatures are shown in Fig. 1a. The standard (1 1 0) peak of 2θ for $Ba_{0.6}Sr_{0.4}TiO_3$ ceramic (PDF Number: 34-0411) is at 31.892° (λ = 1.54178 nm). The (1 1 0) peak of 2θ in Fig. 1a is at 31.981, 31.829 and 31.887 for the BST thin films deposited at substrate temperature 650 °C, 680 °C and 700 °C, respectively. Therefore, $Ba_{0.6}Sr_{0.4}TiO_3$ thin films and ceramics have the same cubic perovskite structure at room temperature. The crystallinity and grain size of the films increase with rising substrate temperature. In Fig. 1b the (1 1 0) peaks of 2θ for BST350 are larger than the peak of BST700 about 0.7°. The orientation factor based on Lotgering's theory [11] is defined as

$$F = \frac{P - P_0}{1 - P_0}, \quad P = \frac{\sum I_{(a\,b\,c)}}{\sum I_{(h\,k\,l)}}$$

where F denotes the orientation factor with respect to the reference plane ($a\ b\ c$), where $a\ b\ c$ represents the Miller indices, P means the ratio of the sum of intensities of the interested family of plane ($a\ b\ c$) to the sum of all reflections for the textured thin film, and P_0 stands for the equivalent ratio for ceramic powder of the target with random orientation. The orientation factor of plane (2 1 0) of the thin films deposited at substrate temperature of 650 °C, 680 °C and 700 °C is 0.6598, 0.6637 and 0.7389, which shows that the crystallinity and

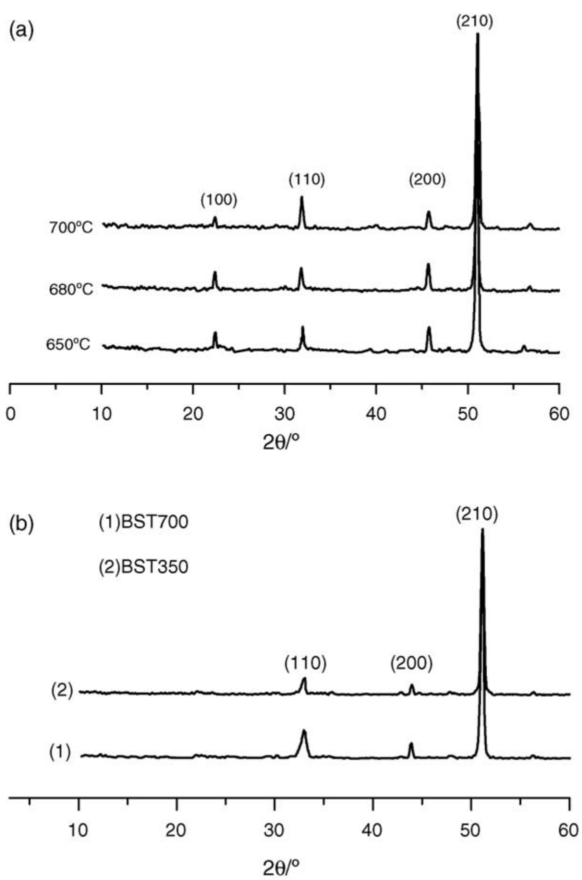

Fig. 1. XRD patterns of BST thin films deposited (a) in oxygen pressure of 0.01 Pa at different substrate temperatures and (b) deposited directly at 700 °C, and deposited at 350 °C then annealed at 700 °C.

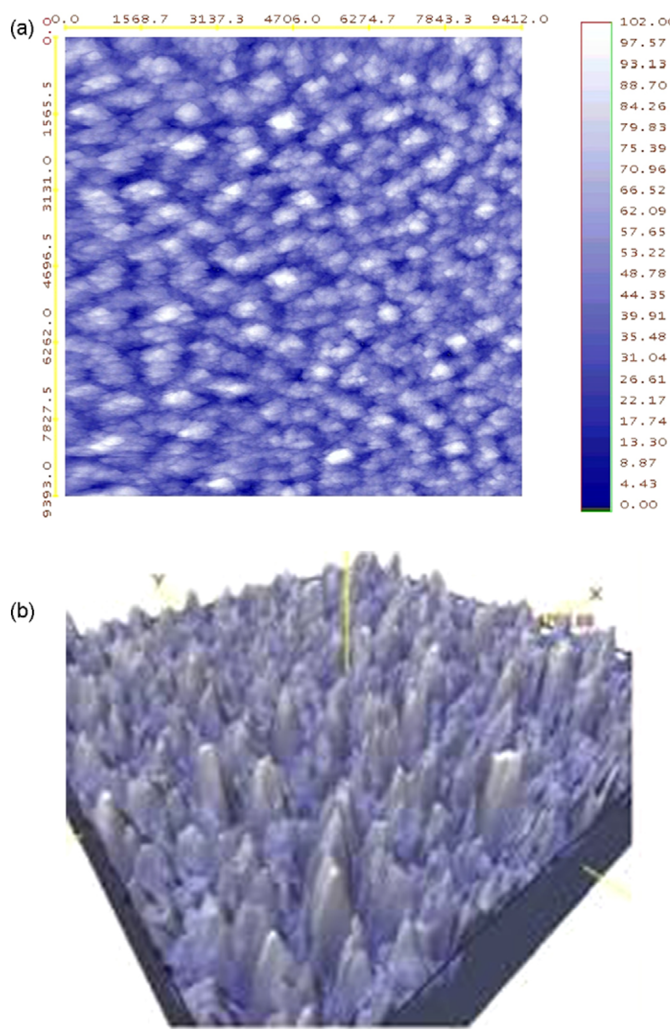

Fig. 2. (a) Two-dimensional and (b) corresponding three-dimensional AFM imagines of BST thin film deposited directly at 700 °C.