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Pulsed laser deposition BTS thin films: The role of substrate temperature

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Abstract

$BaSn_{0.15}Ti_{0.85}O_3$ (BTS) thin films were deposited on $Pt/Ti/SiO_2/Si(1\ 0\ 0)$ substrate by pulsed laser deposition and the effects of substrate temperature on their structure, dielectric properties and leakage current density were investigated. The results indicate that the substrate temperature has a significant effect on the structural and dielectric properties of the BTS thin films which exhibit a polycrystalline perovskite structure if the substrate temperature ranges within 550–750 °C. The dielectric constant and loss tangent of the BTS thin films deposited at 650 °C are 341 and 0.009 at 1 MHz, respectively, the tunability is 72.1% at a dc bias field of 400 kV/cm, while the largest figure of merit (FOM) is 81.1. The effect of the substrate temperature on the leakage current of the BTS thin films is discussed.

Keywords: A. Films; B. Grain size; C. Dielectric properties; E. Capacitors

1. Introduction

With the growing demand for compatibility with integrated circuit technology, interest in the processing of ferroelectric thin films has been increased many fold in the recent years [1–4]. Lead-based ferroelectric materials, especially Pb(Zr, Ti) O_3 (PZT) thin films have been widely studied and used due to their excellent properties [5,6]. However, environmental issues may ultimately require the replacement of these lead-based materials and thin films in electronic components [7]. Therefore, it is important to develop lead-free ferroelectric thin films. $BaTiO_3$-based thin films as perovskite ferroelectric materials are used widely in dielectric, piezoelectric, and electro-optic applications [8,9]. Among them, Barium tin titanate Ba $(Sn_xTi_{1-x})O_3$ (BTS) is a solid solution compound that exhibits dielectric and ferroelectric properties depending on the specific composition and temperature. In addition, the nonlinear dielectric property of $Ba(Sn_xTi_{1-x})O_3$ makes it very attractive for the future tunable microwave device components, such as phase shifters, filters, varactors, delay lines, etc [10–12].

Different techniques are employed to grow ferroelectric thin films, including sol–gel processing [13], molecular beam epitaxy (MBE) [14], magnetron sputtering [15], metal-organic chemical vapor deposition (MOCVD) [16], chemical solution deposition (CSD) [17], spray pyrolysis [18], pulsed laser deposition (PLD) [12,19], and plasma enhanced chemical vapor deposition (PECVD) [20]. Among them, PLD provides several advantages for the growth of multi-component ferroelectric thin films. The composition of films grown by PLD is quite close to that of the target, and it is true even for a multi-component target. In addition, the surface of the ferroelectric thin films grown by PLD can be very smooth [19,21], which is advantage for microwave device components.

In the PLD process, the properties of the ferroelectric thin films are known to be strongly dependent on the prepared parameters such as substrate temperature, oxygen pressure and film thickness. To determine the growth conditions of BTS ferroelectric thin films by PLD is thus essential for future applications. Substrate temperature is one of crucial parameters for the crystal growth of thin films and has a significant effect on the mobility and kinetic energy of adatoms on the substrate [22], as well as electrical properties of the thin films.

In this paper, $BaSn_{0.15}Ti_{0.85}O_3$ (BTS) thin films were deposited on Pt-coated Si substrates by using the PLD process.

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To obtain large tunability and low dielectric loss, the effects of substrate temperature on phase structure, surface topography, dielectric properties and leakage current density are investigated in detail.

2. Experimental procedure

The $BaSn_{0.15}Ti_{0.85}O_3$ (BTS) ceramic targets were prepared by a conventional solid-state reaction process using raw materials $BaCO_3$ (99.99%), TiO_2 (99.99%) and SnO_2 (99.99%). The starting materials were weighed and mixed according to the stoichiometric mole ratio of the BTS and milled for 6 h. Then the mixed powders were pressed into the disks and sintered at 1100 °C for 12 h. Sintered BTS pellet was used as a target for laser ablation and the thin films were deposited on Pt/Ti/SiO_2/Si (1 0 0) substrates. The substrates were cleaned in an ultrasonic bath with alcohol for 30 min. The substrates were fixed at an on-axis distance of 6 cm from the target. The laser radiation was impinged on the target at 45° with respect to normal in a dynamic flow of oxygen. The deposition was done in oxygen pressure of 15 Pa. Before irradiations, the deposition chamber is evacuated down to a base pressure of $3.0\times10-^{4}$ Pa. The BTS thin films were deposited on the substrate at different temperatures of 450 °C, 550 °C, 650 °C, and 750 °C, respectively. After deposition, the thin films were cooled to room temperature at the same oxygen pressure. All of the thin films with a typical thickness of about 500 nm obtained from the step height measurement instrument.

The crystal structure of the films was characterized by X-ray diffraction using a (XRD DX-2700, Fangyuan) system equipped with a Cu-Kα radiation source (1.542 Å) in the diffraction angle range of 20–60° (increment 0.02°). Thin film morphologies were performed by the atomic force microscopy (AFM) using a Nanoscope Multimode 8 (Bruker, Santa Barbara, CA, USA). The thickness of the thin films was measured by Alpha-Step D-100 profilometer (KLA-Tencor, California, USA). For electrical measurement, Au top electrodes with 0.2 mm in diameter were patterned by lift-off process to form the metal-insulator-metal type capacitors. The dielectric properties, tunability were measured at room temperature by impedance analyzer (Agilent 4294 A, Santa Clara, California, USA). The *P–E* hysteresis loops and leakage current of the films were carried out with a ferroelectric testing system (Precision Workstation, RADIANT, USA) at room temperature.

3. Results and discussion

Fig. 1 shows the XRD patterns of the BTS thin films deposited at various substrate temperatures on Pt/Ti/SiO_2/Si(1 0 0) substrates. The XRD patterns exhibit that the structure of the BTS thin films is strongly dependent on substrate temperature. No peaks due to the crystalline phase could be detected in the BTS thin films deposited at 450 °C which indicates that the thin films are amorphous in nature. The characteristic peaks of the main BTS phases emerge with substrate temperature increase to 550 °C, and possess the polycrystalline perovskite structure [23]. The diffraction peaks

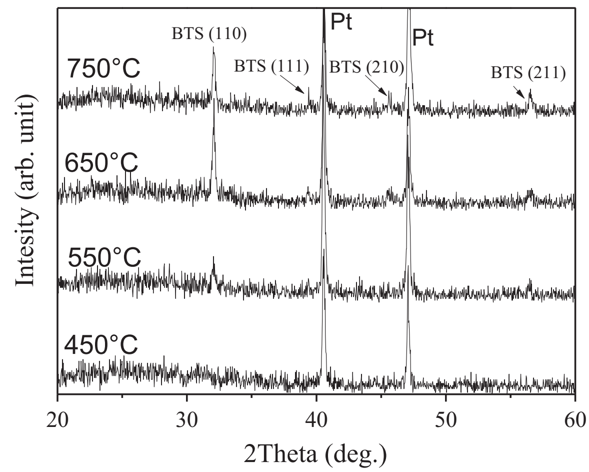

Fig. 1. XRD patterns of BTS thin films deposited on Pt/Ti/SiO_2/Si(1 0 0) with different substrate temperature.

intensity is enhanced obviously when the substrate temperature up to 650 °C. However, with further increase of substrate temperature up to 750 °C, the diffraction peak turns to lower. These can be explained as follows. For the BTS thin films deposited at low substrate temperature ($\leq$550 °C), the evaporated particles are not enough energy to move on the surface of thin films, and there exist lots of defects in the thin films, which influence the nucleation and growth of the thin films, thus lead to the poor crystalline quality. The excess of substrate temperature ($\geq$750 °C) might induce defects in the BTS thin films, which interfere with the nucleation and growth of the thin films, thus lead to the degradation of the crystalline quality. The BTS thin films possess the best crystalline quality when the substrate temperature is 650 °C.

In order to attain the detailed structure information, the grain size along the (1 1 0) plane according to the Scherrer's formula can be calculated [24].

$$D=\frac{0.9\lambda}{\beta\ \cos\ \theta} \tag{1}$$

where D is the mean grain size, λ is the wave length of Cu Kα radiation (1.542 Å), β is full width at half-maximum (FWHM) of the (1 1 0) peak and θ is the Bragg diffraction angle. We have corrected β by considering the effect of instrumental broadening. According to Eq. (1), the D values are 36 nm, 45 nm and 39 nm for the BTS thin films deposited at 550 °C, 650 °C, and 750 °C, respectively. For the BTS thin films deposited at 650 °C, the grain size reaches its maximum due to its smallest FWHM. The results above indicate that there is an optimum substrate where the films show relative better nano-crystallite.

The micrographs of atomic force microscopy (AFM) of the BTS thin films deposited at various substrate temperatures were shown in Fig. 2. The surface morphologies of the BTS thin films deposited at different substrate temperatures are altered. As shown in Fig. 2a, the surface morphology of thin films deposited at 450 °C is an incompact structure, which indicating the amorphous nature of thin films. For the BTS thin