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Dielectric and Ferroelectric Properties of $Ba_{1-x}Sr_xTiO_3$ Thin Films Prepared by Pulsed Laser Deposition

S.Y. Wang, B.L. Cheng*, Can Wang, W. Peng, S.Y. Dai, Z.H. Chen

Institute of Physics, Chinese Academy of Sciences, Beijing 100080, P.R. China

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Abstract. In this work, BST (x=0.7, 0.5, 0.3) films have been deposited onto Nb-$SrTiO_3$ substrate with pulsed laser deposition. The crystal structure and surface morphology have been characterized by X-ray diffraction (XRD) and atomic force microscopy, respectively. XRD results revealed that in certain elaboration condition the films were aligned along (00l) direction, normal to the substrate surface. The dielectric loss, relative dielectric permitivity, and polarization of BST films strongly depended on Sr content at room temperature. The tunability of relative dielectric permitivity of BST films exhibited strongly dependence on Sr content, and BST-0.5 shows the maximun K (K = tunability/loss) value.

Introduction

Recently, solid solutions of barium titanate and strontium titanate, $Ba_{1-x}Sr_xTiO_3$ (BST) have been extensively studied because of their potential applications for various microelectronic devices. BST possesses a wide range of relative dielectric permitivity varying from a few hundred to thousands depending on Ba/Sr ratio, grain size, and temperature [1,2]. In addition, BST exhibits a large tunability of the dielectric permitivity upon application of a dc electric field near Curie temperature. These properties make BST as an attractive material for storage capacitors in next-generation dynamic random access memories and also in electrically tunable microwave devices, such as tunable filters, phase shifters, etc.

Thin films of BST offer additional advantages of lightweight, compactness, lower processing temperature, lower operating voltage, and compatibility with semiconductor processing technology. To achieve high dielectric permitivity and low dielectric loss tangent (tanδ), BST film is usually grown epitaxially on single-crystal substrates such as MgO, $LaAlO_3$ (LAO), and Nb-doped SrTiO3 (STN) [3-5]. The choice of Ba/Sr ratio in BST dielectric layer largely depends on intended operating temperature, as the maximun dielectric permitivity and tunability is typically achieved near the Curie temperature, which varies with Ba/Sr ratio. However, such extrapolation from bulk to thin film is not trivial because of significantly different stresses and oxygen vacancies, which films might possess. In most epitaxial oxide thin films, distortion of unit cell caused by stresses or oxygen vacancies usually cause changes in the electric properties of the films. Pulsed laser deposition (PLD) provides unique advantages for multicomponent oxide films because it easily reproduces the stoichiometry of the target in the deposited films.

For measurement of dielectric and electric properties of BST thin film, a metal (Pt, Ag, Cr, and Au) is widely used as electrode to fabricate a metal-insulator-metal (MIM) capacitor [2-5]. However, there exist some problems with the metal, including etching difficulty and high oxygen permeability, which permits oxygen diffusion from perovskite oxide thin films through the metal electrode to the substrate and surface [6]. The resultant oxygen vacancy formation accelerates the degradation of the dielectric properties of oxide thin films. Nb-doped $SrTiO_3$ (STN) has some potential to be adopted for electrode material due to epitaxial growth of BST thin film on it with high dielectric property [3, 7]. In this work, we report a study of structures and dielectric properties of BST (x=0.7, 0.5, 0.3) films

* Corresponding author. Electronic mail: blcheng@aphy.iphy.ac.cn (Bo Lin Cheng)

grown on STN.

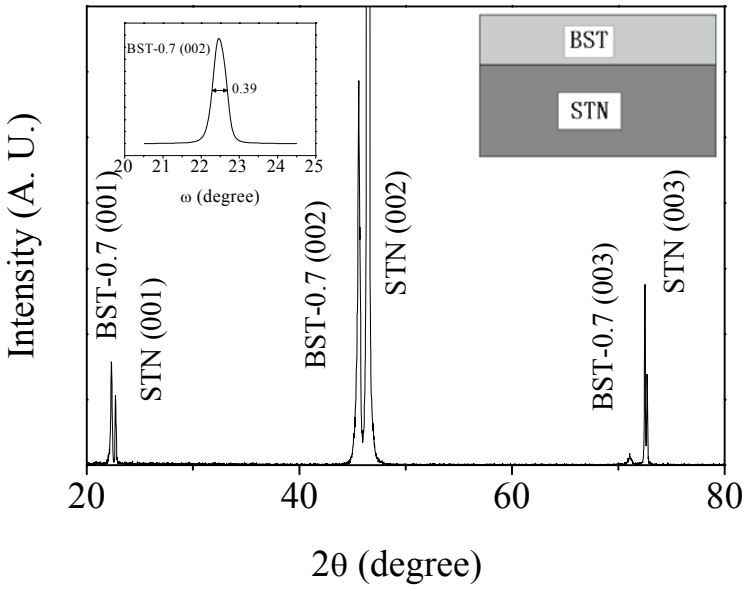

Fig. 1. X-ray 2θ scans of the $Ba_{0.7}Sr_{0.3}TiO_3$ film on Nb-doped $SrTiO_3$. The inset shows the schematic diagram of heterojunction.

Results and discussions

BST films were deposited with pulsed-laser deposition on (001)-STN with thickness of 0.5 mm. To eliminate other deposition variables, we kept the same conditions for film preparations: oxygen pressure of 2 Pa, a deposition temperature between 780 and 800°C, and distance between the target and the substrate of 40 mm. A laser with energy density of about 2 J/cm^2 was focused on the ceramic target of $Ba_{1-x}Sr_xTiO_3$ (x=0.7, 0.5, 0.3). After the deposition, the films were cooled to room temperature in an oxygen pressure of 2×10^4 Pa. The BST films were measured to be about 600 nm by a surface profile measuring system (DEKTAK III, USA).

The epitaxy and lattice parameters of thin films were examined and deduced by X-ray diffraction. X-ray 2θ scans of the $Ba_{0.7}Sr_{0.3}TiO_3$ film on Nb-doped $SrTiO_3$ are shown in Fig. 1. The insets show the schematic diagrams of heterojunctions. Fig. 1 shows θ-2θ XRD scans and rocking curve measurements of BST film on STN. Only (00l) peaks appear in the θ-2θ scans, indicating the film has its c-axis normal to the substrate surface. The rocking curve measurement of (002) reflections reveals that full width at half maximum is 0.39˚. These results suggest that the BST thin film is basically c-orientated single crystal. The out-of-plane lattice parameters for BST films, calculated from the (002) peak position of the BST θ-2θ scans, decrease from 3.977 Å to 3.962 Å, and 3.947 Å with increasing Sr content.

Surface morphologies of BST films were observed with Nano Scope III atomic force microscopy (AFM). Fig. 2 shows the typical AFM image of $Ba_{0.5}Sr_{0.5}TiO_3$ film. The surface morphology did not show dependence on Sr content. The root-mean-square surface roughness (R_{rms}) of thin films is about 0.33 nm. These smooth film surfaces can provide excellent adhesion with Ag top electrodes in MIM type capacitor test structures.

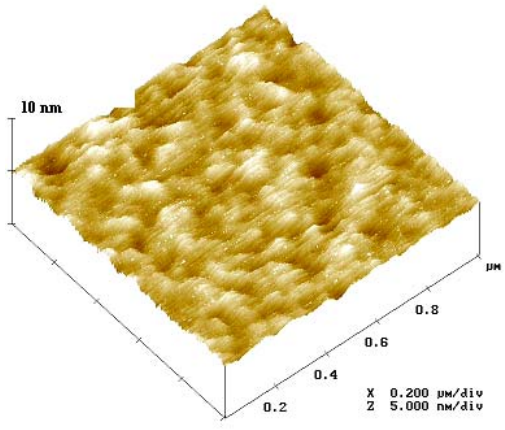

Fig. 2. AFM image of $Ba_{0.5}Sr_{0.5}TiO_3$ film. X: 0.2 μm/div, Y: 0.2 μm/div, Z: 5.0 nm/div

Dielectric measurements of BST films were con- ducted at room temperature on MIM capacitor. Ag top electrodes with a diameter of 0.2 mm were deposited under the pressure of 1×10^{-3} Pa. The relative dielectric permitivity of BST films is calculated from the capacitance using following equation: $\varepsilon_r = C_p\, t / \varepsilon_0\, A$, where C_p is the measured capacitance (F), ε_0 free space permittivity value (8.85×10^{-12} F/m), A capacitor area (m^2) and t the thickness (m) of BST film. Fig. 3 shows the variation of dielectric permitivity of BST films as a function of frequency (1-100 kHz). The BST films show a dependence on Sr content, and BST-0.5 exhibits the maximum permitiviy at 100 kHz, as summarized in Table 1.

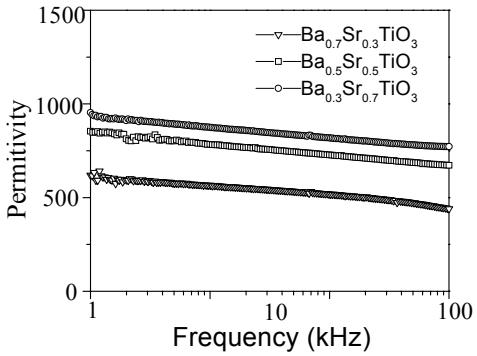

Fig. 2. The relative dielectric permitivity of the Ag/BST/STN capacitors as a function of frequency.

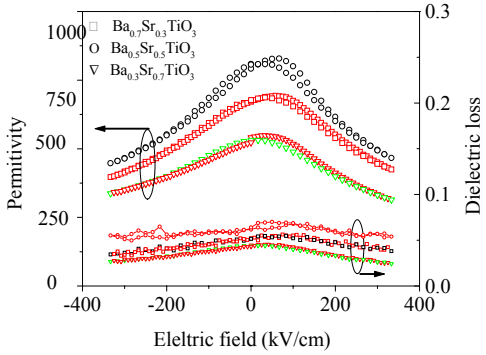

Fig. 3. The relative dielectric permitivity and dielelctric loss of the Ag/BST/STN capacitor as a function of frequency.

Fig. 4 shows electric field dependence of permitivity and dielectric loss tangent of BST films on STN substrate at room temperature. The measurement was carried out on MIM capacitor by using a small ac signal of 50 mV at 100 kHz. In the measurement, the applied electric field scanned from positive 330 kV/cm to negative one. From this figure, we can see the permitivity and dielectric loss of BST films were strongly reduced by applied electric field. The variation of permitivity of the BST films was measured in terms of the parameter of tunability, $[\varepsilon_r(0\text{ bias}) - \varepsilon_r(330\text{ kV/cm})]/\ \varepsilon_r(0\text{ bias}) \times 100\%$. As shown in Tab.1, BST-0.5 film exhibits the highest tunabiltiy (≈45%) at room temperature. At the same time, the dielectric loss also exhibits tunabiltiy with applied electric field. Low dielectric loss is critical for microwave tunable device. For optimum device performance, one need both the highest tunability and minimal dielectric loss. A figure of merit K(K=tunability/loss) is employed to evaluate the performance of BST films. As shown in Tab. 1, BST-0.5 exhibits the highest K value.

It is suggested that the dielectric loss in a ferroelectric film originates from impurity and oxygen vacancies, uniform and local strain, and interfacial effect [3]. Johnson's phenomenological theory could show the permittivity and dielectric loss of BST films decrease with the increasing electric field [8].

Tab. 1. Dielectric and ferroelectric properties of $Ba_{1-x}Sr_xTiO_3$ films

Sample	$2P_r$ ($\mu C/cm^2$)	ε_r (100 kHz)	Tunability (%) (330kV/cm)	Loss (zero bias)	K
BST-0.7	11.6	690	41.2	0.052	7.9
BST-0.5	5.2	805	45.2	0.042	10.8
BST-0.3	2.8	533	36.8	0.043	8.6

A ferroelectric property of BST film affects strongly the dielelctric constant and dielelctri loss in film. Fig. 4 shows polarization versus electric field (P-V) hysteresis of BST films. The P-V hysteresis was measured with RT6000S ferroelectric test system in virtual ground model at room temperature. Remnant polarization 2Pr of BST film decreases with increasing Sr content in $Ba_{1-x}Sr_xTiO_3$ films, as summarized in Tab. 1. It is worth to note that hysteresis loops is not symmetry as a function of electric field. One possible reason for that is the asymmetrical space charge near two interfaces between BST film and electrode [9]. The detailed mechanism of such asymmetry needs further investigation.

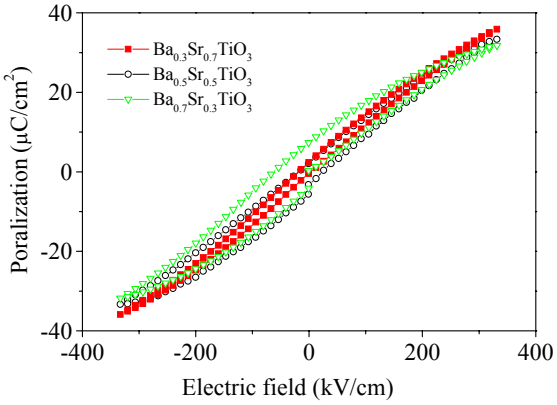

Fig. 4. Ferroelectric hysteresis loops of the Ag/BST/STN capacitors with different x.

Summary

Thin films of $Ba_{1-x}Sr_xTiO_3$ were deposited onto Nb-$SrTiO_3$ substrate with pulsed laser deposition. With special deposition conditions, all BST films were aligned along (00l) direction, normal to substrate surface. The dielectric loss, permitivity and polarization were strongly influenced by the ratio of Ba/Sr, and BST-0.5 exhibits the maximum permitivity and tunabiltiy at room temperature. The BST-0.5 film exhibits the maximum K (K = tunability/ loss) value, indicating that it is more suitable for use in tunable devices at room temperature.

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