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Dielectric enhancement of $BaSrTi_{1.1}O_3/BaSrTi_{1.05}O_3/BaSrTiO_3$ multilayer thin films prepared by RF magnetron sputtering

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Abstract

$BaSrTi_{1.1}O_3/BaSrTi_{1.05}O_3/BaSrTiO_3$ multilayer (BSTM) thin films and $BaSrTiO_3$ (BST) thin films were deposited on $LaNiO_3$ (LNO)/SiO_2/Si substrates by radio frequency (RF) magnetron sputtering at substrate temperature of 400 °C, respectively. X-ray diffraction (XRD), scanning electron microscopy (SEM) and atomic force microscopy (AFM) investigations revealed that all the films have uniform and crack-free surface morphology with a perovskite structure. The dielectric constant of the BSTM thin films was increased and dielectric loss was decreased compared with those of uniform BST thin films. The dielectric constant of 420, dielectric loss of 0.017, and dielectric tunability of 38% were achieved for the BSTM thin films.

Keywords: A. Films; C. Dielectric properties; D. $BaTiO_3$ and titanates

1. Introduction

Perovskite barium strontium titanate (BST) has excellent dielectric properties. As a result, BST thin films have attracted considerable interest for practical use such as high density dynamic random access memories (DRAM) capacitors [1,2] and tunable microwave devices [3,4]. In recent years, embedding passive components into printed circuit board or package substrate is becoming a key technology for system-in-package. To produce embedded thin film capacitors, ferroelectric materials can be used as the dielectric layer. Among various ferroelectrics, BST has been paid much attention worldwide [5,6]. For either DRAM capacitor or embedded capacitor, the dielectric layer is required to have a high dielectric constant, a high dielectric nonlinearity and a low dielectric loss. The dielectric properties at microwave are strongly affected by many factors, such as films thickness, grain size, Ba/Sr ratio, and film stress. Recently, deviation of the (Ba+Sr)/Ti ratio from the stoichiometric value of unity is reported to have a dramatic effect on the dielectric properties. Stemmer et al. [7] fabricated $(Ba_xSr_{1-x})Ti_{1+y}O_{3+z}$ thin films by chemical vapor deposition. Reasonable film behavior is generally achieved up to $y=0.15$, which greatly exceeds the solubility of excess Ti in bulk BST, of approximately $y<0.001$. Jia et al. [8] deposited the epitaxial $Ba_{0.6}Sr_{0.4}TiO_3$ films with TiO_2-doped, and found that by controlling the amount of TiO_2 added to BST reduced the dielectric loss of the films. However, in those papers, the dielectric constant and tunability of the films also reduced with the (Ba+Sr)/Ti ratio increased. In order to optimize the reasonable tunability and dielectric loss of the BST tunable devices, a multilayer structure was discussed. Zhao et al. [9] prepared $BaTiO_3/BaSrTiO_3/SrTiO_3$ multilayer thin films, and found that the dielectric constant was significantly increased. Peng et al. [10] fabricated $Ba_{0.6}Sr_{0.4}Ti_{1+y}O_3/Ba_{0.6}Sr_{0.4}TiO_3/Ba_{0.6}Sr_{0.4}Ti_{1+y}O_3$ multilayer structure films. They reported that the multilayer structure was beneficial to lowering the dielectric loss and enhancing the tunability.

The perovskite oxide $LaNiO_3$ (LNO) electrode has attracted the special attention of several research groups because of a good lattice match and structural compatibility

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with ferroelectric films, which are promising for improving crystal growth and electrical properties of films. It is well-known that LNO has the advantage of decreasing the leakage current and polarization fatigue of the ferroelectric thin films. Chu and Lin [11] showed that the BST films had crystallization with the LNO electrode even at a low temperature of 200 °C. However, the dielectric constant of BST films was moderate and the crystallinity needs to be further improved. There are few studies on the multilayer thin films fabricated by RF magnetron sputtering technology at low temperature.

In the present work, BSTM thin films and BST thin films were prepared by RF magnetron sputtering technology on LNO/SiO_2/Si substrate at substrate temperature of 400 °C. The microstructure and dielectric properties of the BSTM thin films and BST thin films were investigated.

2. Experiment

LNO bottom electrode about 80 nm in thickness was deposited on SiO_2/Si substrate using the RF magnetron sputtering. Then, the sample was annealed at 750 °C for 1.5 min in rapid thermal annealing (RTA) furnace (Mila-3000, ULVAC-RIKO, Japan) to pre-crystallized LNO film. The heating ratio of RTA was 40 °C/s. After that, 380 nm-thick BST thin films were grown on LNO/SiO_2/Si substrates at 400 °C by the RF magnetron sputtering. The BSTM thin films were deposited on LNO/SiO_2/Si substrates by the RF magnetron sputtering using three stoichiometry targets in turn: $BaSrTiO_3$, $BaSrTi_{1.05}O_3$ ($BST_{1.05}$), $BaSrTi_{1.1}O_3$ ($BST_{1.1}$). Each target was sputtered for an hour, the total thickness of BSTM thin films is about 380 nm. The other sputtering parameters of BST films and BSTM films were kept under the same conditions and summarized in Table 1. Finally, the samples were annealed at 800 °C for 15 s in RTA and at 700 °C for 1.5 h in a tube furnace and then cooled naturally. The heating ratio was 20 °C/s for RTA and 2 °C/min for conventional heating process.

The crystal structure of the BSTM films and BST films was analyzed by X-ray diffraction (XRD). The surface morphology was determined by a scanning electron microscopy (SEM) and atomic force microscopy (AFM). For electrical measurements of the BSTM films and BST films, top gold (Au) electrodes of 0.4 mm diameter were deposited through a shadow mask onto the BSTM films and BST films by sputtering. The dielectric properties and *C*–*V* characteristics of Au/BSTM/LNO and Au/BST/LNO capacitors were measured by using an Agilent 4294 A impedance analyzer at room temperature.

Table 1
Deposition conditions of LNO, BST and BSTM thin films.

Item	LNO	BST	BSTM
Target-substrate distance (mm)	50	50	50
Substrate temperature (°C)	300	400	400
RF power (W)	60	70	70
Rate of Ar:O_2	4:1	4:1	4:1
Working pressure (Pa)	2	3.5	3.5

3. Results and discussion

Fig. 1 shows the XRD patterns of the BSTM thin films and BST thin films. As shown in Fig. 1, all films are polycrystalline with a perovskite structure. The peaks of (110) is much higher than the others, indicating clearly a preferred (110) orientation in these films.

The microstructure, such as crystallinity and grain size, is one of the key parameters determining the electrical properties of dielectric thin film capacitor. The surface morphologies and cross-sectional images of BSTM film and BST film was analyzed using SEM and AFM. Fig. 2 shows the SEM and AFM micrographs of BSTM film and BST film. It can be observed that all films show compact, smooth and uniform morphologies, no cracks and pits can be found on the surface of the films. The BST films have larger grain than the BSTM films. For BSTM films, the average grain size determined from the SEM is about 50 nm and the root-mean square (RMS) roughness is around 3.5 nm by the AFM analysis. For BST films, the average grain size is about 60 nm and the RMS roughness is around 3.8 nm. The faintly visible interfaces can be observed inside the compositionally graded thin films in the cross-sectional SEM images. The BSTM films possess a polycrystalline structure and the BST films possess a columnar structure.

Fig. 3 shows the dielectric constant (ε_r) and dielectric loss (tan δ) of BSTM thin films and BST thin films as a function of frequency at room temperature. The relative dielectric constant was calculated from the capacitance data using the classical formula of parallel-plate capacitors. From Fig. 3 we can see that the dielectric constant of all the films decreases with increasing frequency. The ε_r of

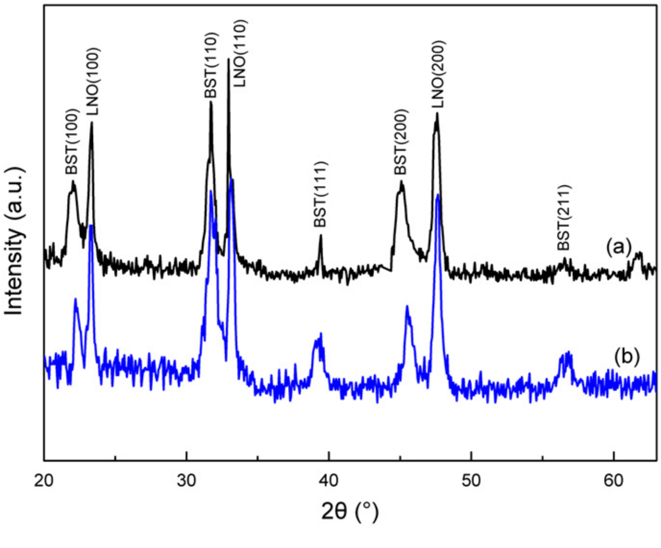

Fig. 1. XRD patterns of the samples: (a) BSTM thin films and (b) BST thin films.