Improvement of dielectric loss tangent of Al_2O_3 doped $Ba_{0.5}Sr_{0.5}TiO_3$ thin films for tunable microwave devices

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Al_2O_3 doped $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films, with different Al_2O_3 contents, were deposited on (100) $LaAlO_3$ substrates by the pulsed laser deposition technique to develop agile thin films for tunable microwave device applications. The dielectric properties of Al_2O_3 doped BST films were determined with a nondestructive dual resonator near 7.7 GHz. We demonstrated that the Al_2O_3 doping plays a significant role in improving the dielectric properties of BST thin films. The Al_2O_3 doping successfully reduced the dielectric loss tangent ($\tan\delta$) from 0.03 (pure BST) to 0.011 (Al_2O_3 doped BST). Reduction in the loss tangent also leads to reduction in the dielectric constant and dielectric tunability. Our results showed that the BSTA4 film remains tunability=15.9%, which is sufficient for tunable microwave devices applications. Consequently, the Al_2O_3 doping improved the figure of merit (K) for the BST films from $K=7.33$ (pure BST) to $K=14.45$ (Al_2O_3 doped BST). © *2004 American Institute of Physics.* [DOI: 10.1063/1.1638615]

I. INTRODUCTION

It is a challenging task for microwave communication researchers to develop tunable microwave devices. One approach is to use a ferroelectric thin film as a buffer layer, and tune the dielectric constant (ϵ_r) of the ferroelectric thin film by applying an electric field. Hence, the working frequency of the microwave devices can be tuned accordingly.[1–4] The foremost candidate for a hybrid multilayer thin film system involves $Ba_{1-x}Sr_xTiO_3$ (BST), where x is the concentration of the constituent, which can be tuned as well. An additional advantage of BST is the Curie temperature (T_C) of BST dependence on x. Therefore, the working temperature of the tunable microwave devices can be tuned as well.[5–8] Unfortunately, the dielectric loss ($\tan\delta$) of the BST is normally high, being around 0.03 for $x=0.5$ in our present study. The reported values of $\tan\delta$ for BST from different research groups are varied. In general, the values of $\tan\delta$ are too high for BST to be of practical use in microwave tunable devices. It is a general belief that $\tan\delta$ values have to be lower than 0.01 for BST thin films to be useful in microwave devices.[9–11]

In the present study, we chose to address the problem of dielectric loss by doping the BST with a low loss oxide.[12–14] Hence, Al_2O_3 is chosen as a dopant for its low microwave dielectric loss.

II. EXPERIMENT

A BST target with 2.5 cm diam was prepared using $BaTiO_3$, $SrTiO_3$ powders with ratio 1:1, via conventional ceramic processing. The $BaTiO_3$ and $SrTiO_3$ powders were mixed and calcined at 950 °C for 1 h before they were compacted and sintered at 1350 °C for 4 h. The Al_2O_3 doped BST thin films were deposited on (100) $LaAlO_3$ single crystal substrates with a size of $10\times5\times0.5$ mm^3 by pulsed laser deposition (PLD) with a KrF excimer laser at a repetition rate of 5 Hz, and the average pulse energy was 250 mJ. The BST target, with Al_2O_3 plates on its surface, was employed in the film deposition. The deposition of Al_2O_3 doped BST films was carried out at a substrate temperature of 650 °C and an oxygen pressure of 0.2 mbar for 45 min. The post-deposition annealing was done in the PLD chamber at the same temperature but with a higher oxygen pressure of 1.0 atm. The optimum distance between substrate and target was 4.5 cm for this temperature and pressure. The content of Al_2O_3 in the deposited Al_2O_3 doped BST films was controlled by the coverage area of Al_2O_3 over the BST target. BST film without doping was also deposited for comparison. The films produced from the targets with 10%, 20%, 30%, and 40% Al_2O_3 coverage of the BST target were abbreviated to BSTA1, BSTA2, BSTA3, and BSTA4. The Al_2O_3 content in the films was characterized by Rutherford backscattering (RBS) analysis in combination with proton induced x-ray emission (PIXE). In our experiment, only the total aluminum

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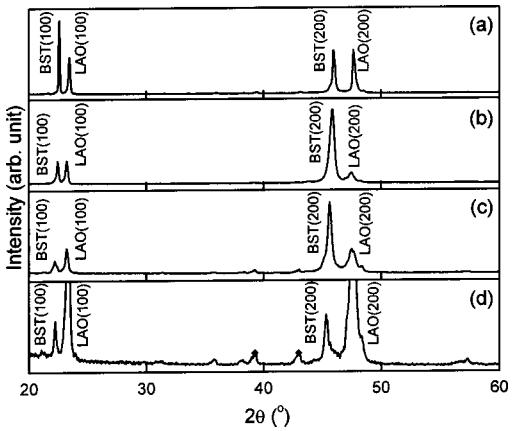

FIG. 1. XRD patterns of the Al_2O_3 doped BST films on $LaAlO_3$ substrates: (a) BSTA1, (b) BSTA2, (c) BSTA3, and (d) BSTA4.

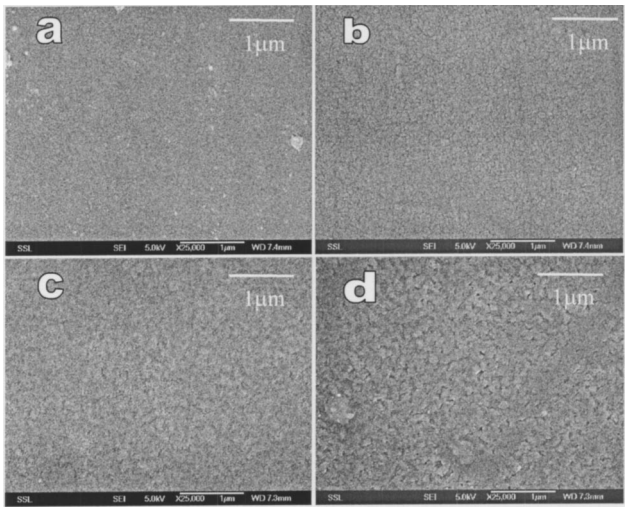

FIG. 2. Surface morphology of the Al_2O_3 doped BST films on $LaAlO_3$ substrates: (a) BSTA1, (b) BSTA2, (c) BSTA3, and (d) BSTA4.

(Al) content can be quantified from the PIXE data. Therefore, the relative concentration of Al_2O_3 in BSTA1:BSTA2:BSTA3:BSTA4 was found to be 1:4:6:9.

The structural phase composition and crystallization of the Al_2O_3 doped BST thin films were determined by x-ray diffraction (XRD), using a Philips PW 1729 type x-ray diffractometer with Cu $K\alpha$ radiation. Surface morphology was examined by scanning electron microscopy (SEM), using a JEOL JSM-6340F type field emission scanning electron microscope.

The dielectric properties of Al_2O_3 doped BST films in terms of the dielectric constant (ϵ_r) and dielectric loss tangent ($\tan\delta$) were measured by a homemade nondestructive microstrip dual-resonator method at room temperature and microwave frequency ~7.7 GHz.[15,16] The microstrip dual-resonator, patterned on a TMM10i microwave substrate, consists of two planar half-wavelength resonators coupled through a gap of 36 μm. The film under test was placed on top of the microstrip circuit, covering the gap between two microstrip resonators. The dielectric constant (ϵ_r) and dielectric loss tangent ($\tan\delta$) of the films were derived from the resonant frequencies f_1, f_2 and the corresponding quality factor Q_1, Q_2, of the microstrip dual resonator. The method was verified by measuring the dielectric properties of the $LaAlO_3$ single crystal substrate, with $\epsilon_r = 21$ and $\tan\delta = 2 \times 10^{-5}$, which has been characterized for $LaAlO_3$ single crystal substrates. In the study of the electric field dependence of the Al_2O_3 doped BST thin films, a maximum dc voltage of 2.1 kV was applied through two electrode pads on the microstrip circuit board across a gap of about 2.6 mm, corresponding to a maximum electric field of ~8.1 kV/cm.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of the deposited thin films. It reveals that all the films have a single phase perovskite structure. The d_{k00} of BST increase linearly with the Al_2O_3 content, as shown by the position of the XRD peak for the BST ($k00$), which shifted to the lower angular side in Fig. 1. In our present study, the BST is a psuedocubic structure. The increase in d_{k00} of BST also indicates the increase in the lattice constant, and can be additional evidence of the incorporation of Al_2O_3 into BST. The Al_2O_3 was not detected by XRD due to the relatively small amount of Al_2O_3 compared with BST. However, for the BSTA4 film, two unidentified peaks were observed at $2\theta \approx 39.39°$ and $2\theta \approx 43.01°$ (marked ◆ in Fig. 1). These peaks are attributed to a second phase caused by the excessive Al_2O_3 content.

Figure 2 displays the surface morphology of the Al_2O_3 doped BST thin films by SEM. The films exhibited a dense microstructure, which was greatly modified by Al_2O_3 doping. The SEM micrographs in Fig. 2 showed that the surface roughness increases with the increase of Al_2O_3 content. Furthermore, the thicknesses of the thin films were estimated as 500 nm on average from a cross-sectional SEM image.

The dielectric constant (ϵ_r) of the Al_2O_3 doped BST film as a function of the applied electric field is shown in Fig. 3. The dielectric constant for both doped and undoped BST films has been characterized and listed in Table I. The pure BST film has $\epsilon_r = 1622$, which is higher than the doped films, meanwhile the dielectric constants for BSTA1, BSTA2, BSTA3, and BSTA4 films are 1387, 1311, 950, and 870, respectively. The ϵ_r of Al_2O_3 doped BST films decrease with

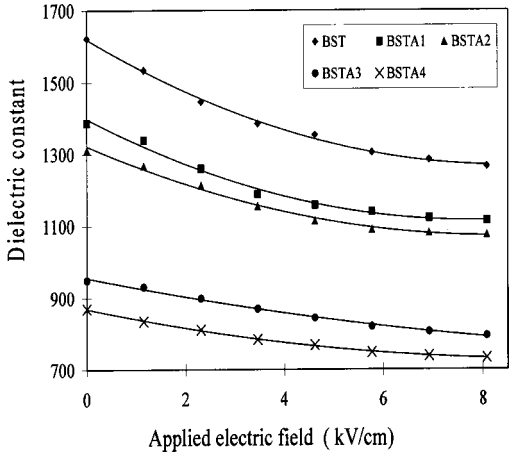

FIG. 3. Dielectric constant of the Al_2O_3 doped BST films on $LaAlO_3$ substrates as a function of applied electric field.

TABLE I. Dielectric properties of the Al_2O_3 doped BST films at microwave frequency ~7.7 GHz.

Thin films	ϵ_{r_0}	tan δ (for 0 bias)	Dielectric tunability (%) (8.1 kV/cm)	Figure of merit (K) (8.1 kV/cm)
BST	1622	0.030	22.0	7.33
BSTA1	1387	0.021	19.7	9.38
BSTA2	1311	0.015	17.9	11.93
BSTA3	950	0.012	16.4	13.67
BSTA4	870	0.011	15.9	14.45

increasing Al_2O_3 content. The high ϵ_r values are due to the fact that all thin films are epitaxially *c*-axis oriented, which are indicated by the (100) and (200) peaks in the XRD patterns. The highly *c*-axis oriented films provide a stronger polarization direction compared to randomly oriented samples, and tend to form a concentrated polarization, which results in a higher dielectric constant.[5,13] Figure 4 depicts the plot for tan δ of Al_2O_3 doped BST films as a function of the applied electric field. The tan δ decreases by 0.030, 0.021, 0.015, 0.012, and 0.011, corresponding to the BST, BSTA1, BSTA2, BSTA3, and BSTA4 samples, respectively. The BSTA4 film (tan δ=0.011) is more applicable for fabrication of tunable microwave devices compared with the BST film (tan δ=0.03), due to its low tan δ, which enhances performance.[9–11]

It is known that tunable microwave device applications require a high dielectric tunability and low dielectric loss. The dielectric tunability was calculated by the formula

$$\frac{\epsilon_{r_0}-\epsilon_{r_b}}{\epsilon_{r_0}}\times 100\%, \tag{1}$$

where ϵ_{r_0} and ϵ_{r_b} represent the dielectric constant value at zero applied electric field and the maximum applied electric field, respectively. The dielectric properties of Al_2O_3 doped BST films are listed in Table I. There is inevitably a tradeoff when improving the dielectric loss tangent of ferroelectric thin films. Thus, reduction in the dielectric loss tangent leads to reduction in dielectric constant and dielectric tunability as a consequence of Al_2O_3 doping. We found that the dielectric tunability is decreased from 22.0% (BST) to 15.9% (BSTA4). However, the dielectric tunability for the BSTA4 film is still acceptable for tunable device applications. In tunable microwave device applications, the figure of merit (K) is usually used to compare the quality of ferroelectric films. The figure of merits (K) can be define as

$$K=\left[\frac{\text{tunability}}{\tan\delta}\right]=\left[\frac{(\epsilon_{r_0}-\epsilon_{r_b})/\epsilon_{r_0}}{\tan\delta}\right]. \tag{2}$$

One can notice that the K factor is proportional to Al_2O_3 content, as given in Table I in which $K=7.33$ (BST) and $K=14.5$ (BSTA4). Although the effects of Al_2O_3 doping reduced the dielectric constant, dielectric loss tangent, and dielectric tunability, it also doubled the figure of merit for the BST films, as compared with the K factor for BSTA4 and BST. The K factor showed that the Al_2O_3 doping succeeded in improving the dielectric properties of BST films, enhancing their suitability in tunable microwave device applications with their low loss and high K factor.

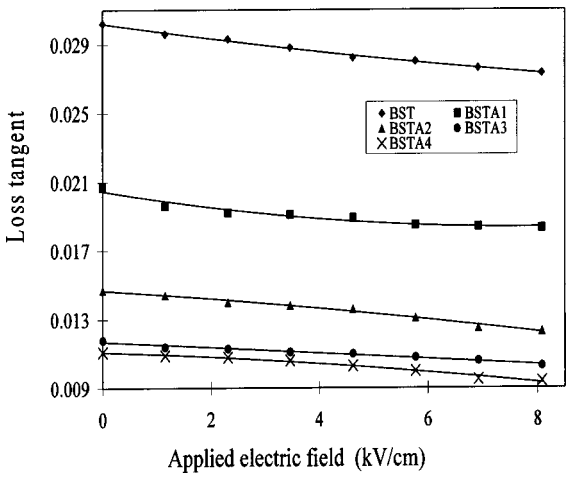

FIG. 4. Loss tangent of the Al_2O_3 doped BST films on $LaAlO_3$ substrates as a function of applied electric field.

IV. SUMMARY

We demonstrated that highly *c*-axis oriented Al_2O_3 doped BST films could be deposited on (100) $LaAlO_3$ single-crystal substrates by PLD technique. The Al_2O_3 substitution into BST films is observed to significantly modify the microstructure of the films. We concluded that the Al_2O_3 doping significantly improved the dielectric loss tangent of the BST thin films. The dielectric loss tangent has been reduced from 0.03 (BST) to 0.011 (BSTA4) with Al_2O_3 doping. Identical to the dielectric loss tangent, the dielectric constant and dielectric tunability were also reduced, but nevertheless, the dielectric tunability for BSTA4 film remains 15.9% and is sufficient for tunable microwave device applications. As a result, Al_2O_3 doping improved the K factor significantly, with comparison between $K=14.5$ (BSTA4) and $K=7.33$ (BST). Our results showed that the dielectric properties of BST ferroelectric thin films can be readily modified with Al_2O_3 doping to fit the requirements of microwave device applications.

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