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# Improvement of dielectric loss tangent of $\text{Al}_2\text{O}_3$ doped $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ thin films for tunable microwave devices

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$\text{Al}_2\text{O}_3$  doped  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$  (BST) thin films, with different  $\text{Al}_2\text{O}_3$  contents, were deposited on (100)  $\text{LaAlO}_3$  substrates by the pulsed laser deposition technique to develop agile thin films for tunable microwave device applications. The dielectric properties of  $\text{Al}_2\text{O}_3$  doped BST films were determined with a nondestructive dual resonator near 7.7 GHz. We demonstrated that the  $\text{Al}_2\text{O}_3$  doping plays a significant role in improving the dielectric properties of BST thin films. The  $\text{Al}_2\text{O}_3$  doping successfully reduced the dielectric loss tangent ( $\tan \delta$ ) from 0.03 (pure BST) to 0.011 ( $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  doping improved the figure of merit ( $K$ ) for the BST films from  $K=7.33$  (pure BST) to  $K=14.45$  ( $\text{Al}_2\text{O}_3$  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 ( $\epsilon_r$ ) of the ferroelectric thin film by applying an electric field. Hence, the working frequency of the microwave devices can be tuned accordingly.<sup>1–4</sup> The foremost candidate for a hybrid multilayer thin film system involves  $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$  (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.<sup>5–8</sup> 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.<sup>9–11</sup>

In the present study, we chose to address the problem of dielectric loss by doping the BST with a low loss oxide.<sup>12–14</sup> Hence,  $\text{Al}_2\text{O}_3$  is chosen as a dopant for its low microwave dielectric loss.

## II. EXPERIMENT

A BST target with 2.5 cm diam was prepared using  $\text{BaTiO}_3$ ,  $\text{SrTiO}_3$  powders with ratio 1:1, via conventional ceramic processing. The  $\text{BaTiO}_3$  and  $\text{SrTiO}_3$  powders were mixed and calcined at 950 °C for 1 h before they were compacted and sintered at 1350 °C for 4 h. The  $\text{Al}_2\text{O}_3$  doped BST thin films were deposited on (100)  $\text{LaAlO}_3$  single crystal substrates with a size of  $10 \times 5 \times 0.5 \text{ mm}^3$  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  $\text{Al}_2\text{O}_3$  plates on its surface, was employed in the film deposition. The deposition of  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  in the deposited  $\text{Al}_2\text{O}_3$  doped BST films was controlled by the coverage area of  $\text{Al}_2\text{O}_3$  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%  $\text{Al}_2\text{O}_3$  coverage of the BST target were abbreviated to BSTA1, BSTA2, BSTA3, and BSTA4. The  $\text{Al}_2\text{O}_3$  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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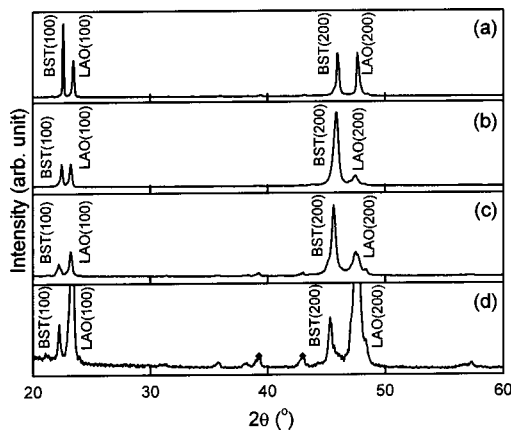


FIG. 1. XRD patterns of the  $\text{Al}_2\text{O}_3$  doped BST films on  $\text{LaAlO}_3$  substrates: (a) BSTA1, (b) BSTA2, (c) BSTA3, and (d) BSTA4.

(Al) content can be quantified from the PIXE data. Therefore, the relative concentration of  $\text{Al}_2\text{O}_3$  in BSTA1:BSTA2:BSTA3:BSTA4 was found to be 1:4:6:9.

The structural phase composition and crystallization of the  $\text{Al}_2\text{O}_3$  doped BST thin films were determined by x-ray diffraction (XRD), using a Philips PW 1729 type x-ray diffractometer with  $\text{Cu K}\alpha$  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  $\text{Al}_2\text{O}_3$  doped BST films in terms of the dielectric constant ( $\epsilon_r$ ) and dielectric loss tangent ( $\tan \delta$ ) were measured by a homemade nondestructive microstrip dual-resonator method at room temperature and microwave frequency  $\sim 7.7$  GHz.<sup>15,16</sup> The microstrip dual-resonator, patterned on a TMM10i microwave substrate, consists of two planar half-wavelength resonators coupled through a gap of  $36 \mu\text{m}$ . The film under test was placed on top of the microstrip circuit, covering the gap between two microstrip resonators. The dielectric constant ( $\epsilon_r$ ) 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  $\text{LaAlO}_3$  single crystal substrate, with  $\epsilon_r = 21$  and  $\tan \delta = 2 \times 10^{-5}$ , which has been characterized for  $\text{LaAlO}_3$  single crystal substrates. In the study of the electric field dependence of the  $\text{Al}_2\text{O}_3$  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  $\sim 8.1$  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  $\text{Al}_2\text{O}_3$  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 pseudocubic structure. The increase in  $d_{k00}$  of BST also indicates the increase in the lattice constant, and can be additional evidence of the

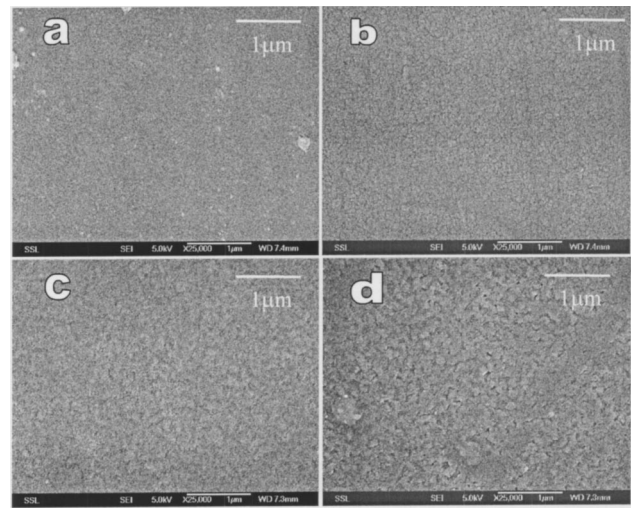


FIG. 2. Surface morphology of the  $\text{Al}_2\text{O}_3$  doped BST films on  $\text{LaAlO}_3$  substrates: (a) BSTA1, (b) BSTA2, (c) BSTA3, and (d) BSTA4.

incorporation of  $\text{Al}_2\text{O}_3$  into BST. The  $\text{Al}_2\text{O}_3$  was not detected by XRD due to the relatively small amount of  $\text{Al}_2\text{O}_3$  compared with BST. However, for the BSTA4 film, two unidentified peaks were observed at  $2\theta \approx 39.39^\circ$  and  $2\theta \approx 43.01^\circ$  (marked  $\diamond$  in Fig. 1). These peaks are attributed to a second phase caused by the excessive  $\text{Al}_2\text{O}_3$  content.

Figure 2 displays the surface morphology of the  $\text{Al}_2\text{O}_3$  doped BST thin films by SEM. The films exhibited a dense microstructure, which was greatly modified by  $\text{Al}_2\text{O}_3$  doping. The SEM micrographs in Fig. 2 showed that the surface roughness increases with the increase of  $\text{Al}_2\text{O}_3$  content. Furthermore, the thicknesses of the thin films were estimated as 500 nm on average from a cross-sectional SEM image.

The dielectric constant ( $\epsilon_r$ ) of the  $\text{Al}_2\text{O}_3$  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  $\epsilon_r$  of  $\text{Al}_2\text{O}_3$  doped BST films decrease with

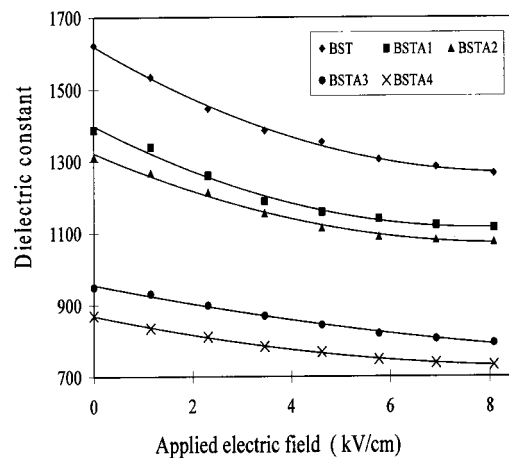


FIG. 3. Dielectric constant of the  $\text{Al}_2\text{O}_3$  doped BST films on  $\text{LaAlO}_3$  substrates as a function of applied electric field.

TABLE I. Dielectric properties of the  $\text{Al}_2\text{O}_3$  doped BST films at microwave frequency  $\sim 7.7$  GHz.

Thin films	$\epsilon_{r_0}$	$\tan \delta$ (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  $\text{Al}_2\text{O}_3$  content. The high  $\epsilon_r$  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.<sup>5,13</sup> Figure 4 depicts the plot for  $\tan \delta$  of  $\text{Al}_2\text{O}_3$  doped BST films as a function of the applied electric field. The  $\tan \delta$  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 ( $\tan \delta=0.011$ ) is more applicable for fabrication of tunable microwave devices compared with the BST film ( $\tan \delta=0.03$ ), due to its low  $\tan \delta$ , which enhances performance.<sup>9–11</sup>

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

$$\frac{\epsilon_{r_0} - \epsilon_{r_b}}{\epsilon_{r_0}} \times 100\%, \quad (1)$$

where  $\epsilon_{r_0}$  and  $\epsilon_{r_b}$  represent the dielectric constant value at zero applied electric field and the maximum applied electric field, respectively. The dielectric properties of  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  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

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

One can notice that the  $K$  factor is proportional to  $\text{Al}_2\text{O}_3$  content, as given in Table I in which  $K=7.33$  (BST) and  $K=14.5$  (BSTA4). Although the effects of  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  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.

#### IV. SUMMARY

We demonstrated that highly  $c$ -axis oriented  $\text{Al}_2\text{O}_3$  doped BST films could be deposited on (100)  $\text{LaAlO}_3$  single-crystal substrates by PLD technique. The  $\text{Al}_2\text{O}_3$  substitution into BST films is observed to significantly modify the microstructure of the films. We concluded that the  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  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,  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  doping to fit the requirements of microwave device applications.

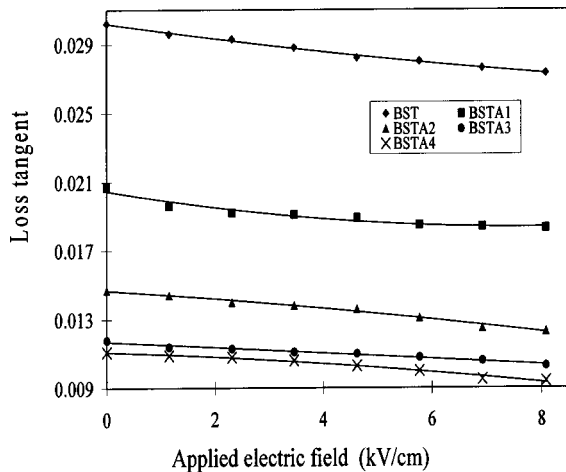


FIG. 4. Loss tangent of the  $\text{Al}_2\text{O}_3$  doped BST films on  $\text{LaAlO}_3$  substrates as a function of applied electric field.

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