Ba$_{0.5}$Sr$_{0.5}$TiO$_3$–Bi$_{1.5}$Zn$_{1.0}$Nb$_{1.5}$O$_7$ composite thin films with promising microwave dielectric properties for microwave device applications

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Crack-free, dense, and uniform Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ (BST)–Bi$_{1.5}$Zn$_{1.0}$Nb$_{1.5}$O$_7$ (BZN) composite thin films were deposited on (100) LaAlO$_3$, (100) SrTiO$_3$, and (100) MgO substrates via a pulsed laser deposition, using a combined target of BST and BZN ceramics. Phase composition and microstructure of the BST-BZN thin films were characterized by x-ray diffraction and scanning electron microscopy. The films, on LAO, STO, and MgO substrates, showed zero-field microwave (≈7.7 GHz) dielectric constants of 471, 435, and 401, dielectric loss tangents of 0.0048, 0.0043, and 0.0037, and dielectric tunabilities of 6.2%, 6.0%, and 5.7% at ≈8.1 kV/cm, respectively. The good physical and electrical properties of the BST–BZN composite thin films make them promising candidates for microwave device applications. © 2004 American Institute of Physics.

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Ferroelectric thin films have been widely considered to be promising candidates for microwave tunable device applications, such as tunable oscillators, phase shifters, and varactors. High dielectric tunability and low dielectric loss are important properties for these applications. Generally, ferroelectrics with Curie temperature below the operating temperature are employed in practical device applications, since the paraelectric state of ferroelectric materials has lower dielectric loss due to the disappearance of hysteresis. Ferroelectric materials, for example, barium strontium titanate (Ba$_{1-x}$Sr$_x$TiO$_3$ or BST), are the most promising candidates for these applications.

The restraint to practical device applications of ferroelectric materials is their dielectric loss tangent. Many efforts have been made to reduce the dielectric loss tangent of ferroelectric materials. It was found that doping of low loss oxides into ferroelectric materials is an effective way to reduce dielectric loss. Various oxides, such as MgO, ZrO$_2$, TiO$_2$, and Al$_2$O$_3$, have been used as additives to lower the dielectric loss tangent of BST thin films. For example, Joshi and Cole prepared MgO-doped BST thin films using metalorganic solution deposition technique. They found that both dielectric loss and insulating characteristics of the doped BST thin films were significantly improved as compared to the undoped case. Other methods, such as, sol–gel process, electrophoretic deposition, and pulsed laser deposition, were also employed to produce MgO-doped BST or MgO-BST composite films with improved dielectric characteristics. The reduced dielectric losses of BST as a result of the addition of the oxides were at the expense of reduction in dielectric constant, as well as dielectric tunability. It was found that Mg occupied BST lattice site at a concentration of 5 mol%. Mg substitution into the BST structure shifted the cubic–tetragonal phase transition peak ($T_C$) to a lower temperature, resulting in a decreased dielectric constant at room temperature. At higher doping level, the excessive MgO mixed with the Mg-substituted BST. The mixing suppressed and broadened the phase transition peak, which also led to a lower dielectric constant. Both effects were also responsible for the reduction in dielectric tunability and dielectric loss. This explanation is also applicable to the effect of other oxides.

The above mentioned oxides are all nontunable and of relatively low dielectric constant. It has been reported that Bi$_{1.5}$Zn$_{1.0}$Nb$_{1.5}$O$_7$ (BZN) thin films have dielectric tunability. Also, BZN demonstrates very low dielectric loss tangent and relatively high dielectric constant. These properties offer us a good opportunity to use BZN as a dopant for improving microwave dielectric characteristics of Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ thin films. In this letter, we report the preparation and characterization of BST–BZN composite thin films, via a pulsed laser deposition (PLD), using a combined BST–BZN target. The BST–BZN thin films were deposited on (100) LaAlO$_3$ (LAO), (100) MgO, and (100) SrTiO$_3$ (STO) single-crystal substrates. Our results showed that the BST–BZN composite thin films demonstrated very low dielectric loss tangent while retaining reasonably high dielectric constant and dielectric tunability, making them excellent candidates for integration into tunable microwave devices. BST and BZN targets with diameter of about 2.5 cm were prepared via the conventional ceramic processing. The combined target used to deposit the BST–BZN composite thin films consists of half BST and BZN. The BST–BZN thin
films were deposited on the repeatedly cleaned LAO, MgO, and STO single-crystal substrates, via a PLD, with a KrF excimer laser. The deposition was carried out for 45 min, at 5 Hz laser repetition frequency, with an energy density of 250 mJ/pulse. The substrate temperature was 650°C and chamber oxygen pressure was 0.2 mbar during the deposition. The distance between substrate and target was 4.5 cm.

Phase composition and crystallization of the BST–BZN thin films were characterized by x-ray diffraction (XRD), using a Philips PW 1729 type x-ray diffractometer with Cu $K\alpha$ radiation. Surface morphology was examined by JEOL JSM-6340F type field emission scanning electronic microscope (SEM).

Microwave dielectric properties of the BST–BZN thin films were measured using a home-made nondestructive microstrip dual-resonator method at room temperature and microwave frequency of $\sim 7.7$ GHz. The microstrip split resonator, formed by a straight microstrip line with a 36 $\mu$m gap in the center, was patterned on a TMM10i microwave substrate. The films were placed on top of the microstrip line, covering the gap. Dielectric constant and loss tangent of the films were derived from the resonant frequency $f_1$, $f_2$ and quality factor $Q_1$, $Q_2$ of the microstrip dual resonator. The method was verified by measuring the dielectric properties of LaAlO$_3$ single crystal substrate, with $\varepsilon_r=21$ and $\tan \delta=2 \times 10^{-3}$. In the study of the electric-field dependence of the BST–BZN 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.

Figure 1 shows the XRD patterns of the BST–BZN composite thin films deposited on different substrates. The BST–BZN thin films on the three substrates demonstrate similar XRD patterns, with three phases, i.e., substrate, BST and BZN, can be found. This means that no reaction between BST and BZN occurred during the deposition process, confirming the effectiveness of our combined target deposition of the BST–BZN composite thin films. We have not tried to use a composite ceramic disk of BST and BZN as a target to deposit BST–BZN thin films because BST and BZN have different sintering temperature. BZN’s sintering temperature is 1300°C. Also, the reaction between BST and BZN at high processing temperature may be unavoidable. It is necessary to mention that the BST perovskite (110) diffraction peak appears in the sample on LAO, whereas almost no such a peak can be found in the films on STO and MgO substrates. That is, the former one is of polycrystalline characteristic, while the latter two are epitaxial. This could be due to the presence of BZN, since pure BST can be easily grown epitaxially on a LAO single-crystal substrate. However, discussion in detail on this issue is not within the scope of the present letter. It is also noted that the (002) peaks of the films on MgO, STO, and LAO are at $\theta=46.18^\circ$, 45.84°, and 45.35°, respectively, meaning that the lattice constant of BST increases with decreasing lattice constant of the substrates.

SEM images of the BST–BZN composite thin films on the three substrates are shown in Fig. 2. It is demonstrated that crack-free, dense, and uniform BST–BZN thin films can...
be readily grown on different substrates. The good physical properties of the BST–BZN thin films are the basic requirement of microwave device fabrications. The grains of the BST–BZN thin film on LAO are equiaxed, while those of the samples on STO and MgO are elongated. The equiaxed grains of the BST–BZN thin film on LAO substrate is one of the characteristics of polycrystalline structure, which is in good agreement with the XRD measurement as discussed above.

The microwave dielectric constant, loss tangent and dielectric tunability of the BST–BZN composite thin films are listed in Table I. The zero-electric field dielectric constants of the BST–BZN films on LAO, STO, and MgO are 471, 435, and 401, with a maximum dielectric tunability of 6.2%, 6.1% and 5.7%, respectively. Their dielectric loss tangent values are 0.0048, 0.0043, and 0.0037, respectively. In comparison, a pure BST thin film deposited on LAO with the same processing parameters has a dielectric constant of 1622, a maximum dielectric tunability of 22% and a loss tangent of 0.031. Relative to the pure BST, all dielectric parameters of the BST–BZN thin films are reduced. The reduced dielectric parameters of the BST thin films can be a result of the presence of BZN phase in BST, which is similar to that observed in the cases of BST doped with other oxides.

From a device impedance matching point of view, ferroelectric thin films for microwave device applications should have a certain value of dielectric constant (~500), a large dielectric tunability (as large as possible) and a low dielectric loss tangent (0.01 or less). If the dielectric constants exceed the required values for planar integrated microwave components, less efficient power transfer will take place in the device, which degrades device performances. High dielectric losses, corresponding to an attenuation of the microwave signal, result in inferior device performance. Specifically, for phase shifter applications, a low dielectric loss tangent is desirable to reduce the insertion loss and, hence, increase the phase shift per decibel of loss. Note that though the maximum electric field applied in our measurement fixture is only ~8.1 kV/cm, the dielectric tunability of the BST–BZN thin films in the present study is comparable to or even better than the reported values. For example, the lowest value of loss tangent was reported by Ngo et al., where Ba0.60Sr0.40TiO3-20 wt% -MgO thick films deposited via an electrophoretic deposition process had a dielectric loss tangent of 0.002. Although this value is only one-half of those in the present study, our BST–BZN thin films have better dielectric tunability than the BST–MgO thick films (8% at 20 kV/cm). Furthermore, high dielectric tunability can be easily realized by increasing the external applied electric field in device fabrications. In this respect, our BST–BZN thin films should be promising candidates for the above mentioned microwave device applications.

We have deposited crack-free, dense, and uniform BST-BZN composite thin films on different single crystal substrates via a PLD. The films, on LAO, STO, and MgO substrates, showed zero-field microwave dielectric constants of 471, 435, and 401, dielectric loss tangents of 0.0048, 0.0043, and 0.0037, and dielectric tunabilities of 6.2%, 6.0% and 5.7% at ~8.1 kV/cm, respectively. In summary, the good physical and electrical properties of the BST–BZN composite thin films make them promising candidates for microwave device applications.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dielectric constant at zero field</th>
<th>Loss tangent at zero field</th>
<th>Tunability (%) at ~8.1 kV/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>BST–BZN/LAO</td>
<td>471</td>
<td>0.0048</td>
<td>6.2</td>
</tr>
<tr>
<td>BST–BZN/MgO</td>
<td>403</td>
<td>0.0037</td>
<td>5.7</td>
</tr>
<tr>
<td>BST–BZN/STO</td>
<td>435</td>
<td>0.0043</td>
<td>6.0</td>
</tr>
</tbody>
</table>

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