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A low-loss BST thin film on initial nucleation layer for micro and millimeter wave tunable phase shifter

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Abstract

Barium strontium titanate, (Ba,Sr)TiO₃ (BST), thin films have been deposited on (100)-oriented MgO substrate by combining preparation of initial layer by Pulsed Laser Deposition and main layer by Metal-Organic-Decomposition method. Films with an initial layer of 20, 30 and 40 nm thickness and final thickness of 400, 650 and 800 nm have been obtained. Physical and dielectric properties of the BST thin films have been characterized from the viewpoint of frequency-agile micro and millimeter circuit applications. The results reveal that $Ba_{0.6}Sr_{0.4}TiO_3$ thin films have a good crystallinity with characteristic orientation that is affected by the deposition conditions of the initial layer. Interdigital capacitor with a gap of 10 µm has been characterized and the dielectric loss and tunability are as low as 0.002–0.004 and 12%, respectively, at frequency of 1 MHz for the applied voltage from \pm 40 to \pm 40 V. At microwave frequencies, classic-shaped coplanar waveguide lines formed on BST/(100) MgO were investigated. A differential phase shift of 18° was obtained at 20 GHz with insertion loss of about –2 dB at 60 V for Au/Cr interconnection. Finally, a three-stage LC-ladder-type phase shifter with variable capacitors of BST thin film has been fabricated considering the experimental results obtained for the classic-shaped coplanar waveguide lines and a maximum phase shift of 40° is obtained at 20 GHz and 60 V. © 2005 Elsevier Ltd. All rights reserved.

Keywords: BST; Sol-gel processes; Dielectric properties

1. Introduction

In the last decade, a strong interest was gathered on the development of tunable dielectric materials for frequency-agile microwave circuit applications.¹ The high dielectric nonlinearity (high-DC-field-dependent dielectric permittivity) of ferroelectric materials has made them promising candidates for tunable microwave applications. Among these materials, strontium titanate, SrTiO₃ (STO), and barium strontium titanate, (Ba,Sr)TiO₃ (BST), are the most promising ferroelectric materials for the development of tunable dielectric microwave circuits. Especially BST shows controllable Curie temperature by changing its chemical composition and a very large change in permittivity value with a small electric field. This feature makes BST one of the most investigated materials for such applications.² Regardless of deposition condition, BST thin films revealed a low dielectric loss that can be further lowered by careful consideration on the deposition parameters, and a promising tunability.

0955-2219/\$ - see front matter © 2005 Elsevier Ltd. All rights reserved. doi:10.1016/j.jeurceramsoc.2005.09.015 Also, from the viewpoint of tunable device fabrication using a ferroelectric material, an improvement in the device performance and a reduction of fabrication cost are expected.

Coating of water-based MOD solution directly on MgO was proved to be very difficult due to small adhesion between the solution and the substrate. Even if this problem was solved by selecting the appropriate deposition conditions, we encountered a difficulty in depositing BST films with a thickness more than 150 nm due to poor adhesion between the MgO substrate and MOD deposited BST film. In order to ease the deposition of water-based BST solution on (1 0 0) MgO substrates, and to have a control over the strain caused by the mismatch between MgO and BST lattice parameters, an intermediate layer (interlayer) of BST having the same chemical composition as the main layer has been deposited by PLD on the MgO substrates. An advantage for this onset is the combination between the easiness of BST deposition by PLD with the deposition on large size substrates and good film uniformity given by MOD deposition technique.

In this paper, BST thin films of 400, 650 and 800 nm thickness have been deposited by a Metal Organic Decomposition (MOD) method on $(1\ 0\ 0)$ MgO substrates on which an initial layer (interlayer) of BST was deposited by Pulsed Laser Deposition (PLD)

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technique. The initial BST layer is 20, 30 or 40 nm in thickness and has the same chemical composition as the main MOD-made BST layer. Tunability and dielectric loss have been measured on interdigital (ID) capacitors and analysis at high frequencies has been performed on classic-shaped coplanar waveguide (CPW) lines. Moreover, a three-stage LC-ladder type phase shifter with variable capacitors of BST film was designed and evaluated.

2. Experimental

The deposition conditions for the intermediate layer are shown in Table 1. Substrate temperature and the temperature and time of BST interlayer annealing have been simultaneously modified to ensure a large change in film properties. Two kinds of interlayer on MgO were prepared. The first, having a thickness of 20, 30 or 40 nm has been deposited on MgO substrates heated at 600 °C and annealed at the same temperature in O₂ atmosphere at a pressure of 200 m Torr for 30 min. The other one, having a thickness of 20 nm, have been deposited on MgO substrates at room temperature and annealed at atmospheric pressure for 1 h at 800 °C in an O₂-rich environment.

After the interlayer deposition and annealing, a 0.06 M Metal-Organic Decomposition (MOD) solution with Ba/Sr ratio of 60/40 was coated to prepare BST films. Ba_{0.6}Sr_{0.4}TiO₃ (BST60) solution has been coated on (100) MgO substrate with the interlayer already deposited and annealed. A good uniformity on the substrate was achieved by spinning for 3 s at 500 rpm followed by another 20 s at 3000 rpm in air. Coating step was followed by drying at 250 °C for 10 min in an oven. After drying, the BST60 films were baked at 450 °C for 10 min in an oven in O2-rich atmosphere to burn the residual organics and to enhance chemical reaction of oxides. The deposition was repeated 10 times and was followed by a heat-treatment (annealing) at 800 °C for 10 min in O₂-rich atmosphere. The entire process was then repeated several times until the desired thickness had been obtained. The final annealing was performed at 800 °C for 20 min in O2-rich atmosphere. The total thickness for the BST films obtained was 400, 650 and 800 nm. Drying temperature of 250 °C and baking temperature of 450 °C were selected to minimize as much as possible any physical defects (such as cracks on the surface of the film) by analysis of differential thermal (DTA) and thermogravimetric (TGA) behavior of BST solution (not shown here). The crystallinity of the BST films obtained has been investigated by X-ray diffractometry (XRD).

In order to investigate the film behavior at microwave frequencies, a series of interdigitated electrodes (IDEs) and coplanar waveguide (CPW) lines have been fabricated on top of BST thin film by using Au/Cr deposition by vacuum evaporation technique, followed by wet-etching to form Au/Cr metal electrode patterns. Microwave frequency measurements



Fig. 1. X-ray diffraction patterns for BST film with interlayer deposited on MgO at 600 $^\circ\text{C}$ and room temperature.

were carried out using a vector network analyzer (Anritsu, 37247C).

3. Results and discussion

The XRD analysis of the BST thin films (Fig. 1) revealed excellent crystallinities and different main orientations for films deposited on interlayers with different deposition conditions. The BST film on the interlayer deposited at 600 °C has a (110) main orientation accompanied by a small (100) [(200)] peak. On the other hand, when the interlayer has been deposited at room temperature, the film has (100) and (110) main orientations, where peak intensities become almost the same. This is caused by the different conditions for the interlayer even though the deposition conditions for the MOD-made main layer remains the same.

In order to design a tunable phase shifter circuit, along with having films with a small dielectric loss, it is very important to know how the dielectric constant of the BST film is changing with an applied voltage. To improve the figure of merit of the phase shifter, it is also necessary to make the variation of the dielectric constant with the applied voltage to be as large as possible. To investigate these points, a series of interdigitated electrodes (IDEs) and CPW transmission lines were fabricated on the BST film. The interdigital capacitors have finger spacing of 10 µm. Measured tunability and dielectric loss at 1 MHz and a maximum applied voltage of 40 V are shown for the two kinds of interlayer in Table 2. For ID capacitors with a finger spacing of 10 µm formed on a 400 nm thick BST film, the tunability decreases in the capacitor on films with the interlayer deposited on MgO substrate at room temperature (Table 2) and the dielectric constant does not change its small values. Increase in the thickness of the interlayer from 20 nm to 30 or 40 nm

| Deposition conditions of BST | interlayer deposition by PLD |
|------------------------------|------------------------------|

Table 1

| Laser frequency (Hz) | Laser power (mJ) | Substrate temperature (°C) | Pressure (m Torr)/gas | Deposition time (min) | Annealing temperature/time |
|----------------------|------------------|----------------------------|-----------------------|-----------------------|----------------------------|
| 5 | 120 | 600 | 200/O ₂ | 10, 15, 20 | 600 °C/30 min |
| 5 | 120 | RT | 200/O ₂ | 10 | 800 °C/1 h |

Table 2

Tunability and dielectric loss for BST interdigital capacitors with an interlayer deposited at 600 $^\circ C$ and room temperature

| Properties | Tunability (%) | Dielectric loss |
|------------------------|----------------|-----------------|
| Interlayer fabrication | | |
| 600 °C | 11.7 | 0.004-0.007 |
| RT + 800 °C | 11.4 | 0.004-0.007 |

The gap of IDE is 10 µm.



Fig. 2. Tunability vs. interlayer thickness for BST films with the same total thickness of 650 nm.

seems to have a negative effect on tunability (Fig. 2). As the value of tunability is originated mainly from the MOD-made BST layer because of the small interlayer thickness, the reason for this behavior is attributed to the relaxation in strain for the MOD-made BST layer. The strain is caused by the lattice mismatch between MgO and BST interlayer film. It is already demonstrated that the highest tunability is obtained for BST thin films in which the strain is present and has an optimum value.² According to this films with a larger strain or less strain have a smaller tunability. Increase in the total BST film thickness induces an increment in tunability as expected (Fig. 3), with the dielectric loss remaining at small values of 0.002-0.005. From these investigations, it is found that the highest variation of the dielectric constant with the applied voltage is obtained when the interlayer thickness was 20 nm, deposited at substrate temperature of 600 °C and annealed at the same temperature



Fig. 3. Tunability vs. total BST film thickness with films having the same interlayer thickness of 20 nm.



Fig. 4. Differential phase shift vs. frequency for a classic-shaped CPW line with width of $60 \,\mu$ m, gap of $10 \,\mu$ m and length of 2.5 mm. (BST film thickness is 600 nm.)

in 200 m Torr pressure of O_2 atmosphere. Also, it can be seen that the relative variation of dielectric constant increases with increased BST total film thickness.

For further analysis at microwave frequencies, therefore, the interlayer film thickness was optimized at 20 nm. Coplanar waveguide lines formed on films with total thickness of 400, 650 and 800 nm have been investigated. When applying d.c. bias voltage of 0-60 V to the input of a CPW line with width of $60 \,\mu\text{m}$, gap of $10 \,\mu\text{m}$ and length of $2.5 \,\text{mm}$, a differential phase shift of 18° was obtained at 20 GHz (Fig. 4) for CPW on 650 nm BST thin films with insertion loss of about -2 dB for Au/Cr interconnection. For comparison, the phase shift obtained for a CPW with the same parameters formed on BST films of 400 nm thickness was 6° when applying the same d.c. voltage. This agrees qualitatively with the BST thickness dependence of tunability in Fig. 3. The figure of merit for the CPW on 650 nm thin BST film was calculated to be $9^{\circ}/dB$. The measured S parameters were found to agree well with those simulated by MicroWave Office ver.6.

Using the experimental data obtained for CPW line and ID capacitor, a three-stage LC-ladder-type phase shifter with variable BST film capacitors has been designed (Fig. 5). The new phase shifter was designed to have a differential phase shift of about 40° at 20 GHz and 60 V. The experimental results indicate a phase shift of 40° , as shown in Fig. 6.



Fig. 5. Electrode pattern of designed three-stage LC-ladder-type phase shifter.



Fig. 6. Differential phase shift vs. frequency for fabricated phase shifter as a parameter of applied d.c. bias voltage.

4. Conclusions

We have successfully obtained a low loss BST thin film on (100) MgO substrate by preparation of initial layer by PLD followed by deposition of the main layer by MOD method. BST films with a thickness of 400, 650 and 800 nm have been made

having an interlayer deposited at 600 °C or room temperature. Low dielectric losses ranging from 0.002 to 0.007 and tunabilities between 11.5 and 12.1% have been found for ID capacitors with a finger spacing of 10 μ m. At 20 GHz, a phase shift of 18° was obtained for a classic-shaped CPW line with width of 60 μ m, gap of 10 μ m and length of 2.5 mm when applying a d.c. bias voltage of 0–60 V. The insertion loss was –2 dB and the figure of merit was calculated to be 9°/dB. The above results were used to design a three-stage LC-ladder-type phase shifter with variable capacitors of BST films where the phase shift at 20 GHz was 40°. Finally, it was found that the new BST film process is very promising for realization of micro and millimeter wave tunable device.

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