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Microwave Dielectric Properties of Ferroelectric $BaTiO_3$ Thin Film

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Abstract. The dielectric properties of c-axis epitaxial $BaTiO_3$ thin film on $LaAlO_3$ are investigated at frequencies of 0.5–30 GHz. For the measurements, interdigital capacitors with the Au/Ti electrode configurations of five fingers pairs that are 15 μm wide and spaced 2 μm apart are prepared by photolithography and lift-off patterning. Finger length varies from 20 to 80 μm. The capacitance of epitaxial $BaTiO_3$ films exhibited no frequency dependence up to 10 GHz with the exception of slightly upward tendency of capacitance in $BaTiO_3$ film with a finger length of 80 μm due to the self resonant frequency at 20 GHz. The Q-factors of the capacitors, defined as $Q = 1/\omega CR$, are decreased up to 10 GHz with increased frequency. At 10 GHz, the $BaTiO_3$ film has a tunability [defined as $k(V) = [C(0) - C(V)]C(0)]$ of 1.5% at 15 V, a loss tangent of ≤ 0.2 at room temperature. The small tunability can be interpreted as a result of in-plane compressive stress of $BaTiO_3$ film exhibiting large dielectric anisotropy. For the improvement of tunability and dielectric loss in the interdigital $BaTiO_3$ capacitor, the tetragonality (c/a) of epitaxial $BaTiO_3$ film and design of interdigital capacitor should be modified.

Keywords: microwave, dielectric, epitaxial, $BaTiO_3$, polar phase

1. Introduction

Ferroelectric perovskite-type oxide thin films such as $Ba_xSr_{1-x}TiO_3$ and $SrTiO_3$ have been intensively investigated for microwave and millimeter-wave applications because of their large nonlinear dielectric property which is defined as the strong dependence of permittivity on electric field. As candidate materials for microwave applications, ferroelectrics of a paraelectric phase have been mainly considered because ferroelectric materials which are in polar phase at room temperature can give rise to additional dielectric loss due to domain-wall motion [1, 2] and the shear sound waves excited by ferroelastic domain walls or piezoelectric domain effect [3]. Also, the intrinsic hysteresis, which appears in DC bias dependence of the capacitance, can impede the application of ferroelectric materials in a polar phase. However, Powles and Jackson [4] observed that the center of the relaxation spectrum by domain-wall motion appears to be at about 3×10^9 Hz and it was investigated by Arlt et al. [3, 5] that relaxation frequency by piezoelectric domains is about 2×10^9 Hz. Therefore, the domain movement and the sound generation excited by piezoelectric effect do not contribute to microwave loss at frequency more than 10 GHz. Also, Boikov et al. [6] reported for the epitaxial $SrRuO_3/BaTiO_3/SrRuO_3$ film structure that Q-factor related to the hysteresis phenomena and tunability of C-V curve can be optimized through the variation of c/a ratio. Moreover, recent studies [7, 8] demonstrated that the $Na_{0.5}K_{0.5}NbO_3$ ferroelectric thin film in a polar phase at room temperature shows the tunability of 13%, high Q-factor ($1/\tan\delta$) = 15–80 at 40 GHz. Nevertheless, little has been published regarding the tunability and Q-factor of ferroelectric materials in a polar phase at microwave frequency range. In this article, the dielectric properties of epitaxial $BaTiO_3$ thin film, which is typical ferroelectric materials with tetragonally distorted perovskite structure at room temperature, was investigated in microwave frequency range of 0.5–30 GHz.

2. Experimental Details

The epitaxial $BaTiO_3$ thin films with 500 nm thickness were deposited on a wafer of (001) single crystal lanthanum aluminate by *RF* magnetron sputtering system operated at a base pressure of about 1×10^{-6} torr. The sputtering was carried out under the working pressure of 10 mtorr using a gas mixture of Ar/O_2 (4:1). An applied *RF* power was 120 W and the substrate temperature was kept at 600°C. Prior to deposition, the target was sputter cleaned during 30 minutes. An array of interdigital capacitors was fabricated on epitaxial $BaTiO_3$ film by photolithography and Au/Ti (80 nm/20 nm) lift-off patterning. The interdigital capacitors have 5 finger pairs with a finger width of 15 μm and a gap of 2 μm. The finger lengths are 20, 40 and 80 μm. The devices were connected via ground-signal-ground (*GSG*) probe to an *HP* 8510 *C* network analyzer in the frequency band 0.5–30 GHz at room temperature. Two port *S*-parameters measured on interdigital capacitors were converted into admittance (Y) parameters. From the relationship $Im(Y) \approx j\omega C$ (where ω is the operating frequency and C is capacitance), the capacitance and Q-factor were measured through de-embedding procedure in a frequency range which the amplitude of capacitance is not affected by resonant frequency.

3. Results and Discussion

Figure 1 shows X-ray diffraction patterns of $BaTiO_3/LaAlO_3$ structures. An X-ray diffractometer θ-2θ scan for the $BaTiO_3/LaAlO_3$ structure reveals peaks only from the $\{001\}$ family planes with a full width half maximum (FHWM) of 1.047°. The absence of any other peaks indicates that the $BaTiO_3$ films are either highly textured or single crystal. Alignment of the film to the substrate was confirmed with pole-figure scan of (110) planes in the $BaTiO_3$ film. The fourfold symmetry corresponding to the $\{110\}$ reflections was observed as shown in Fig. 1, indicating that the $BaTiO_3$ film is single crystal.

Figure 2 shows the frequency dependence of dielectric properties (capacitance and Q-factor ($=1/\tan\delta$)) of an interdigital capacitor fabricated on $BaTiO_3$ film with the finger lengths of 20, 40 and 80 μm. We have observed no or weak frequency dependence of the capacitance in the measured frequency range up to 10 GHz with the exception of a slight increase of capacitance for interdigital capacitor with 80 μm length. This upward tendency of capacitance was originated from the result of *LC*-resonance at 20 GHz, induced by parasitic inductance (L). The expected dielectric relaxation of epitaxial $BaTiO_3$ film wasn't observed over

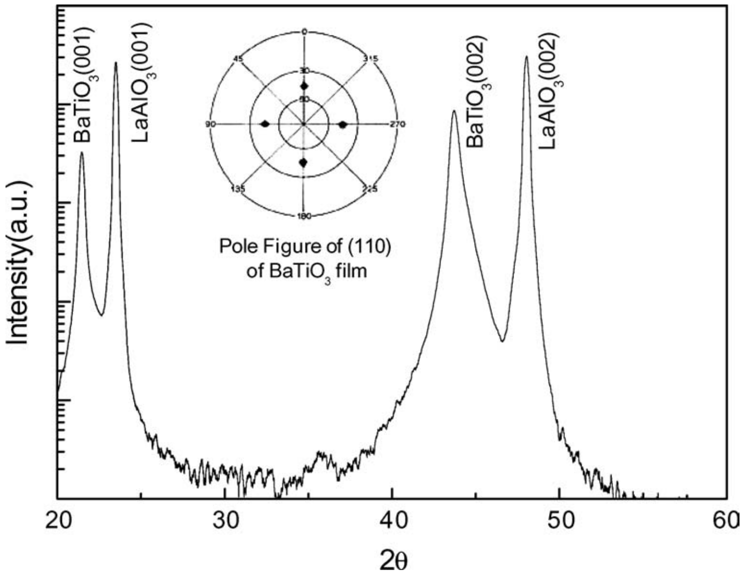

Fig. 1. A typical XRD θ-2θ pattern of $BaTiO_3$ thin film grown on a $LaAlO_3(100)$ substrate. The inset shows the pole figure of (110) plane of $BaTiO_3$film.

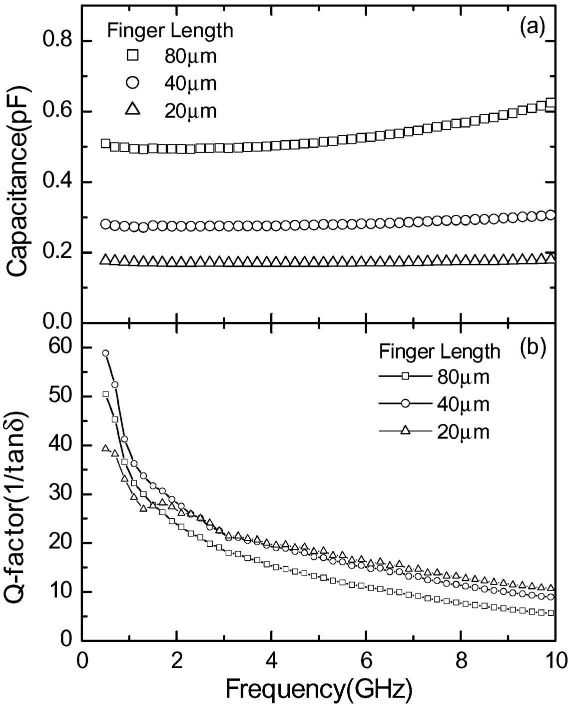

Fig. 2. The frequency dependence of microwave properties measured in interdigital capacitor fabricated on $BaTiO_3$ film, (a) capacitance and (b) Q-factor.

the measured frequency range because in-plane high compressive stress might induce the shift of relaxation frequency and the exact measurement of frequency dependence of capacitance could be interfered with *LC* resonance at 20 GHz. evidently, this *LC* resonance and the quite thin top electrode actually affected dielectric microwave loss. So, it is considered in this work that most of the measured loss tangent was originated from the effect of electrode structure related to its small electrode-thickness and inductance rather than that of the intrinsic dielectric loss (dielectric relaxation) mechanisms.

Figure 3 shows the measured capacitance ($C/C_{\max}$) that is normalized and loss tangent as a function of *DC* bias voltage, taken at 10 GHz from interdigital capacitors fabricated on $BaTiO_3/LaAlO_3$ structure. The tunability, $k(V)$, was calculated by using the equation, $k(V) = [C(0) - C(V)]/C(0)$, where $C(0)$ and $C(V)$ represent the capacitance at zero and a certain dc bias voltage, respectively. The tunability k (15 V) reaches $\sim 1.5\%$ with a loss of ≤ 0.2 at room temperature. The small tunability can be interpreted as a result of in-plane compressive stress of $BaTiO_3$ film exhibiting large dielectric anisotropy. Hyun et al. [9] reported that the in-plane compressive stress in the BST film with an interdigital electrode decrease the ionic displacement along the electric field parallel to substrate and large tunability should exist near the electrode where the electric field is normal to the substrate since the BST film has distorted tetragonal structure with c/a of 1.009 by in-plane compressive stress. In this work, the measured c/a ratio of epitaxial $BaTiO_3$ film was 1.04. This result indicates that the epitaxial $BaTiO_3$ film on $LaAlO_3$ was under compressive strain and had highly distorted tetragonal unit cell with a c-axis normal to

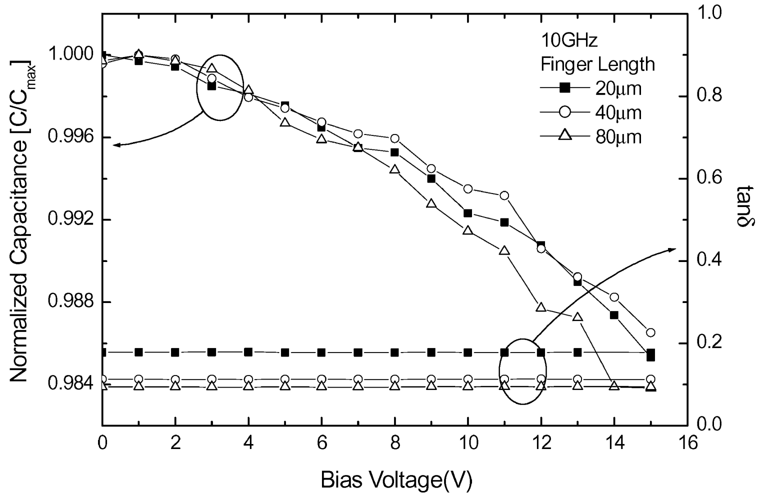

Fig. 3. The capacitance and loss tangent as a function of bias voltage measured in interdigital capacitors fabricated on epitaxial $BaTiO_3$ film with the various finger lengths. The measuring frequency is 10 GHz.

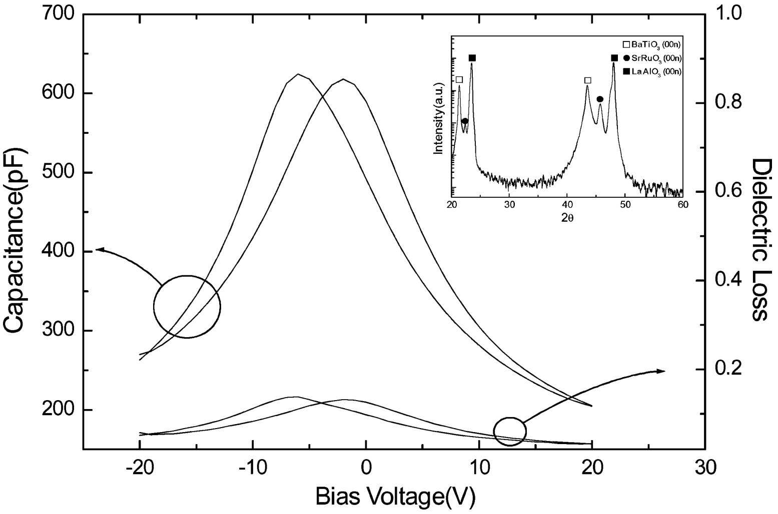

Fig. 4. The capacitance and loss tangent as a function of bias voltage measured in Pt/$BaTiO_3$/$SrRuO_3$ parallel capacitors at 1 MHz. The left inset shows XRD θ-2θ scan for the parallel $BaTiO_3$ capacitor.

the substrate plane. Moreover, it was assumed that the $BaTiO_3$ film under in-plane compressive stress shows tunability only along c-axis perpendicular to the substrate. However, it has been reported that the dielectric constant, ε_{33}, of $BaTiO_3$ single crystal along c-axis is much smaller than ε_{11} and ε_{22} along the a-axis [10] and the lattice elongation along c-axis by high compressive stress in $BaTiO_3$ crystal can lead to a further decrease of the dielectric responses [11, 12]. Also, Boikov et al. [6] reported that a-axis oriented epitaxial $BaTiO_3$ film with c/a of 1.007 have a larger permittivity than c-axis one while the c-axis oriented one should have the largest remnant polarization.

To make sure that the small tunability of interdigital $BaTiO_3$ capacitor can be obtained due to the anisotropic tuning behavior by in-plane stress, parallel capacitor [Pt/$BaTiO_3$ (240 nm)/$SrRuO_3$ (100 nm)/$LaAlO_3$] was fabricated and low frequency characterization (1 MHz) was done by recording the capacitance and dielectric loss as a function of the applied bias. As shown in Fig. 4, there were only (00n) $BaTiO_3$, (00n) $SrRuO_3$ and (00n) $LaAlO_3$ peaks in the X-ray θ-2θ scan and the measured FWHM of the (00n) $BaTiO_3$ film was 1.08°. The tunability k (15 V) of parallel $BaTiO_3$ capacitor at 1 MHz is about 48%, which is much larger than that of interdigital $BaTiO_3$ capacitor. It is considered that this high tunability is originated from removal of possibility of an anisotropic tuning as well as the increased effective strength of *DC* electric field due to the small distance between the top and bottom electrode of parallel capacitor.

Therefore, it is ultimately needed to optimize the Q-factor (defined as tunability/dielectric loss) of interdigital $BaTiO_3$ capacitor through the variation of c/a ratio in epitaxial $BaTiO_3$ as well as the increase of Au electrode and $BaTiO_3$ thickness. The effect of chemical composition on the small tunability was excluded since the stoichiometry of the film agreed well with that of bulk $BaTiO_3$ crystal.

4. Conclusions

We observed the microwave dielectric properties of epitaxial ferroelectric $BaTiO_3$ films grown on (001) $LaAlO_3$ single crystal. The $BaTiO_3$ film with Au/Ti interdigital electrode shows little frequency dependence of capacitance up to 10 GHz although the capacitance of a $BaTiO_3$ film with a finger length of 80 μm slightly increases as a function of frequency because of self-resonance of this capacitor at 20 GHz. The Q-factors of the capacitors are decreased up to 10 GHz as a measuring frequency increases. The $BaTiO_3$ film had a tunability of 1.5% with loss tangent $\leq$0.2 at 10 GHz. It is assumed that the small tunability may be due to in-plane compressive strain. To eliminate the anisotropic tuning behavior and manipulate of hysteresis in tunability for the interdigital $BaTiO_3$ capacitor, c/a ratio of epitaxial $BaTiO_3$ film should be modified. For the improvement of microwave dielectric loss and investigation of exact frequency dependence of capacitance, a new design of interdigital electrode with thick Au/Ti layer is required.

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