Tunable microwave device에 사용될 수 있는 (Ba,Sr)TiO₃ 박막의 유전특성 향상에 관한 연구

Enhanced dielectric properties of (Ba,Sr)TiO₃ thin films applicable to tunable microwave devices

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Abstract

We deposited epitaxial $Ba_{0.6}Sr_{0.4}TiO_3$(BST) films having thickness of 400 nm on MgO(001) substrates, where a 10 nm thick $Ba_{1-x}Sr_xTiO_3$ (x = 0.1 - 0.7) interlayer was inserted between BST and MgO to manipulate the stress of the BST films. Since the main difference of those epitaxial BST films was the lattice constant of the interlayers, we were very successful in controlling the stress of the BST films. BST films under small tensile stress showed larger dielectric constant than that without stress as well as those under compressive stress. Stress relaxation was investigated using epitaxial BST films with various thicknesses grown on different interlayers. For BST films grown on $Ba_{0.7}Sr_{0.3}TiO_3$ interlayers, the critical thickness was about 600 nm. On the other hand, the critical thickness of single-layer BST film was less than 100 nm.

Key Words : $Ba_{0.6}Sr_{0.4}TiO_3$, thin film, strain, pulsed laser deposition, dielectric constant

1. 서 론

$Ba_{1-x}Sr_xTiO_3$ (BST-x) can be used for many electronic devices due to its large dielectric permittivity and electric field dependent dielectric permittivity.[1-3] For example, the large dielectric permittivity makes it attractive for dynamic random access memory devices.[1] On the other hand, the electric field dependent dielectric permittivity makes it a good candidate for electrically tunable microwave devices, which

need large capacitance change ratio under a DC bias.[2,3]

However, it is well known that BST-x thin films show much different physical properties from those of corresponding bulk materials.[4-7] In 1998, Pertsev et al. theoretically showed that misfit strain between a film and a substrate induced a drastic difference between the dielectric properties of thin films and bulk crystals.[8] According to their calculations, $BaTiO_3$ films must show dielectric anomaly at the phase boundary where a polarization component along the measurement direction changes from zero to a finite value.[8]

Recently, several research groups have tried

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to experimentally investigate strain effects on the dielectric properties of BST-x thin films.[9,10] To change strain states of films, the most logical approaches are either variation of substrate materials or variation of deposition conditions, such as oxygen pressure. However, it is very difficult to systematically control the strain states by varying substrate materials due to the limitation of crystal substrates which have the desired lattice constants and thermal expansion coefficients. In addition, variation of deposition conditions such as oxygen pressure can induce oxygen vacancies, which strongly affect dielectric properties through changes in chemical compositions as well as strain states.[11]

In this paper, we report our experimental approach to investigate the strain effects on dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$ (BST-0.4) films. By inserting a very thin BST-x (x = 0.1 - 0.7) interlayer[12] between an epitaxial BST-0.4 thin film and a MgO substrate, we could systematically control the strain states, i.e. the lattice distortion ratio (D = in-plane lattice constant/out-of-plane lattice constant) of the BST-0.4 films. In order to investigate strain relaxations, we also deposited BST-0.4 thin films with different thicknesses on various BST-x interlayers.

2. 실 험

We grow bi-layers of a very thin BST-x (x = 0.1 - 0.7) and a relatively thick BST-0.4 on MgO(001) substrates at 750 ℃ under 200 mTorr oxygen using pulsed laser deposition. Since films could not accommodate large strain imposed by substrates and relax even for very thin films, we used MgO substrates in order to minimize the effects of substrates.[13] [In our early experiments,[14] BST-0.4 films grown on MgO substrates under these conditions showed very close lattice constants to those of a bulk $Ba_{0.6}Sr_{0.4}TiO_3$, similar to others' results.[10,15]]

In order to study the dependence of the lattice

constants of BST-0.4 main layers on the composition of interlayers, we performed x-ray diffraction (XRD) θ-2θ scans using a SIEMENS D5000 x-ray diffractometer. To measure the dielectric properties of the BST films, we also fabricated coplanar capacitors by depositing gold on top of film surface. Each gold electrode had a rectangular shape with a width of 200 m and a length of 2 mm. The separation between gold electrodes for each capacitor was 5 μ m. We measured the capacitance at 1 MHz and room temperature using a HP4194A impedance analyzer. The dielectric constant was calculated using a simplified coplanar waveguide model.[16]

3. 결과 및 고찰

Figure 1 shows the XRD normal (ψ = 90°) θ-2θ scan data of BST-0.4/BST-0.3 and BST-0.4/BST-0.5 bi-layers, where the interlayer BST-x and the main layer BST-0.4 have thicknesses of 10 nm and 400 nm, respectively. Since these data do not show peak separation or extra broadening, we argue that the BST(200) peaks mainly result from BST-0.4 main layers due to the large difference in the film thickness between the interlayer and the main layer (1/40).
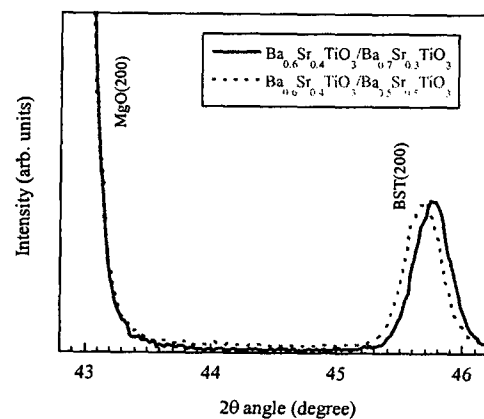

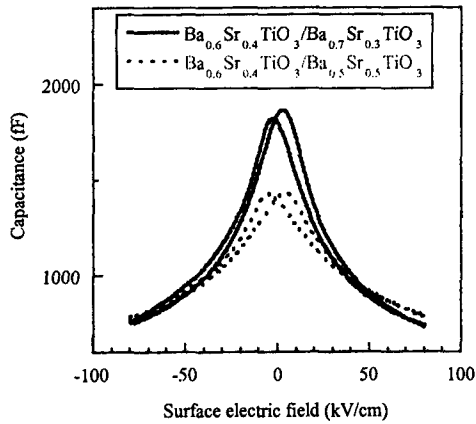그림 1. X 선 회절 패턴.

Fig. 1. X-ray diffraction pattern.

그림 2. 축전 용량 - 전압 거동.

Fig. 2. Capacitance - surface electric field behaviors.

Therefore, from the BST(200) peak positions, we could calculate the out-of-plane lattice constants of the BST-0.4 main layers. Similarly, from the XRD tilted ($\psi = 45°$) $\theta-2\theta$ scan data, we could determine the in-plane lattice constants of the BST-0.4 main layers. The BST-0.4 main layer grown on the BST-0.3 interlayer showed larger in-plane lattice constant (3.971 Å) than out-of-plane lattice constant (3.962 Å). On the other hand, The BST-0.4 main layer grown on the BST-0.5 interlayer showed smaller in-plane lattice constant (3.965 Å) than out-of-plane lattice constant (3.968 Å). Since the interlayer is the only difference of these two BST-0.4 films, we argue that the interlayer plays an important role in determining the lattice constants, i.e. the strain states of the BST-0.4 main layer.

To systematically investigate the effects of the BST-x interlayers on the structural properties of BST-0.4 films, we varied the x in the range from 0.1 to 0.7. As x increased, the out-of-plane lattice constant of the BST-0.4 film increased and in-plane lattice constant decreased. In other words, the lattice distortion ratio (D = in-plane lattice constant/out-of-plane lattice

constant) decreased as x increased.

The dielectric properties of the BST-0.4 are also strongly correlated to the x value of BST-x interlayer. Figure 2 shows the capacitance vs. surface electric field of two BST-0.4 films; one is BST-0.4/BST-0.3 and the other is BST-0.4/BST-0.5. A BST-0.4 film grown on a BST-0.3 interlayer shows larger zero field dielectric permittivity (1180) than that (840) grown on a BST-0.5 interlayer.

4. 결 론

We could systematically control the strain states of a $Ba_{0.6}Sr_{0.4}TiO_3$ film by depositing a very thin $Ba_{1-x}Sr_xTiO_3$ interlayer between the main layer of the $Ba_{0.6}Sr_{0.4}TiO_3$ and a MgO(001) substrate. $Ba_{0.6}Sr_{0.4}TiO_3$ films showed very strong dependence of dielectric properties on the strain states. The strain induced by the MgO substrate was relaxed faster than that induced by an interlayer.

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