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Electrically Tunable Microwave Devices Prepared by rf-Magnetron Sputtering

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Abstract — In this work, we have used the Warren-Averbach method to analyze the x-ray diffraction patterns of several $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (BST) epitaxial thin films deposited via 90° off-axis rf-magnetron sputtering. This method allows for the determination of the coherent strains, incoherent strains, crystallite sizes, and defect densities in these films. Results indicate that for BST (60/40) films, the incoherent strain normal to the substrate is smaller on LAO compared to MgO for films 200 Å – 3000 Å thick. However, the opposite relationship holds for the coherent strains. The thickest films showed dielectric constants of 1350 and 925 on MgO and LAO, respectively (at 1 MHz). All showed dielectric loss on the order of 0.007 with the room temperature tunability being 3 times greater for the films on MgO. In-plane measurements of the coherent strain were also completed, showing good agreement with theoretical predictions of the effects of strain on the dielectric properties.

INTRODUCTION

Electrically tunable microwave devices based on SrTiO_3 (STO) and $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (BST) have been extensively investigated recently [1-3]. In these designs, one takes advantage of the dc electric field tunability of the nonlinear dielectric STO film at cryogenic temperatures. In the case of BST, the operation temperature is dependent upon the Ba/Sr ratio. For example, $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ exhibits the highest tunability near room temperature. For these devices to be practical, it is desirable to have the largest dielectric tunability in combination with the lowest dielectric loss achievable. These dielectric losses originate from various sources, including losses in the bulk of the dielectric film, losses at the interface between the substrate (LaAlO_3 , MgO, etc.) and the dielectric film, and losses at the interface between the film and the electrode. Within the film, such defects as misorientation, grain boundaries, non-stoichiometry, and stress/strain effects can contribute to the loss. Ideally, elimination or minimization of as many of these factors as possible should enhance the dielectric properties of these thin films. The key to this is to understand what influences the presence and magnitude of these factors (i.e., deposition conditions, deposition method, substrate choice, etc.).

EXPERIMENTAL PROCEDURE

Thin film heterostructures were prepared using standard 90° off-axis rf-magnetron sputtering as developed by Eom *et al.* [4]. The high-vacuum system used is equipped with four 2 inch diameter US Gun magnetron sputter sources. In addition, the system is equipped with a load lock/anneal chamber for sample loading/unloading and alternative atmosphere anneals (O_2 , O_3 , etc.) Here, stoichiometric $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ was used as the sputtering target (125 W). The deposition temperature for the BST was typically between 750°C and 800°C , and the sputtering gas composition was a 3:1 ratio of Ar/O_2 at a total pressure of 200 mTorr.

Film structural parameters were determined using conventional x-ray diffraction on a Siemens D5000 diffractometer. $\theta/2\theta$ scans, ω rocking curves and ϕ -scans were completed using this system. Out-of-plane lattice parameters were determined using a Nelson-Riley parameter analysis from the 00 l peaks of the films. These values were confirmed using refinement procedures in the Winjade diffraction pattern analysis software (Materials Data Incorporated, Livermore, CA).

Low frequency dielectric properties were measured on sputter deposited Au coplanar capacitors with a 5 μm gap. The capacitors were fabricated using standard photolithography/liftoff procedures. An HP 4284A LCR meter was used for measurement. The small signal capacitance data reported here were obtained at 100 mV and 1 MHz. In order to estimate the dielectric constant of the BST films, a conformal mapping analytical model was used.

RESULTS AND DISCUSSION

Fig. 1 shows $\theta/2\theta$ and ϕ -scans for BST films deposited on (a) LaAlO_3 (LAO) and (b) MgO. These figures indicate that the BST is deposited epitaxially onto both substrates with only 00 l orientation. LAO can be described with a pseudo-cubic structure having a lattice constant of 3.79 Å. MgO has the rock-salt structure with a lattice parameter of 4.21 Å. BST (60/40) has the cubic perovskite structure with a lattice parameter of 3.965 Å, thus the two substrates chosen here should provide films with significantly different stress/strain states at the interface. Fig. 2 shows atomic force microscopy images

Figure 1: X-ray diffraction patterns for BST (60/40) films deposited on (a) LAO and (b) MgO. The * indicates substrate peaks and $\phi \equiv 0^\circ$ along the substrate [100].

Figure 2: AFM images of BST (60/40) films deposited onto (a) MgO and (b) LAO.

of the surfaces of films deposited on (a) MgO and (b) LAO. Here it can be seen that both substrates result in very smooth films (rms roughness $\approx 3 \text{ \AA}$). The lateral feature sizes are also very similar; on the order of 500 to 800 $\text{\AA}$.

At first glance, therefore, the BST films deposited onto LAO and MgO appear quite similar. There are differences, however. Upon careful examination of the x-ray patterns, one can see a significant difference in the peak shape and position between the two kinds of films. It is important to note that x-ray diffraction can tell us more than simply the orientation of the material under study. The position and breadth of the x-ray peaks hold a significant amount of information which can be very useful. That is, there are two types of structural peak broadening: 1) **Size**: caused by a finite size of regions in the specimen diffracting incoherently with respect to each other, and 2) **Strain**: due to varying displacements of the atoms with respect to their reference positions. In addition to these two, the instrumental contribution to the peak breadth must be accounted for. This is done via measurement of standard materials.

The strain which contributes to the peak broadening will heretofore be referred to as incoherent strain. In addition, there is a coherent strain which is manifested as a shift in the peak position in 2θ with respect to the position determined by bulk material. In terms of a physical description, the coherent strain can be thought of as an overall extension/contraction of the lattice parameter throughout the entire film. The incoherent strain, therefore, is related to a distribution of lattice constants around that peak value. There are a number of methods which can be used to extract the size and strain contributions to the peak breadth. In this work, the Warren-Averbach method was chosen [5].

For this method, as with others, the basis is that the size broadening contribution to the peak breadth is independent of the order of x-ray reflection and the strain broadening contribution is dependent upon the reflection order. The diffraction profile ($P_i(\Delta S)$) is expressed as a Fourier series in terms of the length L perpendicular to the diffracting planes and the deviation $\Delta S = S - S_i$ of the diffraction vector with respect to the origin of the reciprocal space.

$$P_i(\Delta S) = K \sum_{L=-\infty}^{\infty} A_n(L, S_i) \cos(2\pi L \Delta S) + B_n(L, S_i) \sin(2\pi L \Delta S) \quad (1)$$

K is a constant, A_n and B_n are the coefficients of the Fourier series, S_i is, for the i th line profile, the distance of the diffraction vector in reciprocal space with respect to the origin. The sine coefficient can typically be neglected, whereas the cosine coefficient, A_n , can be separated into one part that depends on the size contribution (A_n^s), and another part which is related to the distribution of the average local strain $\varepsilon_L(A_n^d)$. That is,

$$A_n = A_n^s A_n^d \quad (2)$$

The diffraction vector $\mathbf{S}$ can be substituted by the reflection order $d_i^2 = h^2 + k^2 + l^2$ for the i th Fourier coefficient. After separation and a Taylor series expansion, it follows an approximation for small L and d_i :

$$\ln[A_L(d_i)] = \ln[A_n^s(L)] - 2\pi^2 L^2 \frac{d_i^2}{a_3^2} \langle \varepsilon_L^2 \rangle \quad (3)$$

The quantity a_3 represents a fictitious length in reciprocal space. With (3), using at least two measured orders i of reflection, an extrapolation to the zeroth order of d can be made by plotting $\ln A_L(d)$ against d^2 within the Fourier interval of the peaks. This gives the size related Fourier coefficient A_n^s . From the initial slopes of the plots, n represents the number of paircells contributing to the coherently scattering domain and simultaneously the order of the Fourier coefficient. For multiple n , a distribution of the average strain as a function of the length L can directly be obtained.

In this work, both the incoherent and coherent strains were analyzed for out-of-the-plane measurements (001). Fig. 3 shows the results as a function of film thickness. Fig. 3(a) plots the coherent strain (peak shift in 2θ) and fig. 3(b) plots the incoherent strain determined using the Warren-Averbach analysis. It is important to note that the lattice mismatch between BST (60/40) and LAO is -4.3%, while on MgO it is +5.9%. Thus, as described above, one would expect very different strains to be present in these films. In fig. 3, it is clear that the film on the LAO substrate has a larger coherent strain than on the MgO substrate. The opposite relationship holds for the incoherent strain; it is larger for the film on MgO. In addition, there is a clear thickness dependence of the coherent strain.

As mentioned in the introduction, one of the goals of this work is to attempt to understand the sources of the stresses and strains in these films. That is, to determine what deposition variables contribute to these values. As a first step, the effect of increased energetic bombardment conditions was studied. To do this, the substrate was moved 2 mm further into the plasma. The result of this is shown in Fig. 4, where again the coherent and incoherent

Figure 3: Coherent and incoherent strain for BST (60/40) films on LAO and MgO as a function of film thickness.

strains are plotted as a function of film thickness for the increased bombardment conditions. The effect on the magnitude of the coherent strain (peak shift in 2θ) is especially dramatic – there is an order of magnitude increase. Note that a similar thickness dependence of the coherent strain is present for these films, as well. Interestingly, the incoherent strain is relatively unaffected.

Therefore, it is clear that energetic bombardment conditions significantly affect the magnitude of the coherent strain. The thickness dependence in both cases can be explained via the following: For the thinnest films (200 Å – 500 Å), the lattice mismatch induced strain exhibits a larger contribution to the overall strain. That is, since MgO has a larger in-plane lattice constant than BST (60/40), it acts to minimize the out-of-plane lattice extension (via the Poisson effect). Conversely, since LAO has a smaller in-plane lattice constant than BST (60/40), it accentuates any out-of-plane lattice extension. As the films become thicker, the coherent strain (lattice extension) for films on both substrates begin to approach the same values. This indicates that the strain in these films is dominated by bombardment induced defects (since they experienced the same bombardment conditions).

Figure 4: Coherent and incoherent strain for BST (60/40) films deposited on LAO and MgO as a function of thickness. These films were deposited under increased energetic bombardment conditions.

Regarding the incoherent strain, it is not yet entirely clear why the films on LAO show lower values than those on MgO. One possible reason is that the twin transformation in LAO (at $\approx 500^\circ\text{C}$) could relieve some of the incoherent strain upon cooling the sample subsequent to deposition. Another potential explanation is the fact that the films deposited on MgO have a larger absolute value of the lattice mismatch. This could lead to increased inhomogeneous strain in the film.

Finally, x-ray measurements were also completed for in-plane lattice constants, as well (for films in fig. 3). In this case only measurements of coherent strain were completed. Fig. 5 shows these results. As expected, the thinner films show trends consistent with the direction of lattice mismatch. For thicker films, those on MgO approach bulk values, while those on LAO stay relatively constant. Derivations from Devonshire's thermodynamic theory have been completed which describe the effects of these types of strains on the dielectric properties [6]. It was shown that compressive strain (i.e., BST on LAO) results in a slight decrease in the dielectric constant and tunability. Conversely, tensile strain (i.e., BST on MgO) results in a more rapid increase in the dielectric constant and tunability. Here, the thickest films showed dielectric constants of 1350 and 925 on MgO and LAO, respectively (at 1 MHz). All showed dielectric loss on

Figure 5: In-plane coherent strain measured on films deposited under normal energetic bombardment conditions.

the order of 0.007 with the room temperature tunability being 3 times greater for the films on MgO. This shows good agreement with these simple theoretical predictions.

CONCLUSIONS

We have used the Warren-Averbach method to analyze x-ray diffraction patterns of BST (60/40) epitaxial thin films deposited onto LAO and MgO substrates. Coherent and incoherent strains in the films were determined. It was found that the energetic bombardment conditions during deposition strongly influenced the magnitude of the coherent strain.

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