$Sr_xBa_{(1-x)}TiO_3$ THIN FILMS FOR ACTIVE MICROWAVE APPLICATIONS

J.S. HORWITZ, J.M. POND, B. TADAYAN, R.C.Y. AUYEUNG*, P. C. DORSEY, D.B. CHRISEY, S.B. QADRI and C. MULLER**,
Naval Research Laboratory, Washington D.C., *Sachs-Freeman Associates, Landover, MD, **Superconducting Core Technologies, Golden, CO.

ABSTRACT

The dielectric constant, loss tangent and Curie temperature for $Sr_xBa_{(1-x)}TiO_3$ (SBT) thin films with x = 0.2 - 0.8 have been investigated at microwave frequencies. SBT films (0.5 - 3 μm thick) were grown on (100) MgO and $LaAlO_3$ substrates by pulsed laser deposition at substrate temperatures from 850 - 900 °C in 0.35 Torr of oxygen. Deposited ferroelectric films were single phase, highly oriented, and characterized by x-ray rocking curve widths of $\leq 0.5°$. Highly oriented SBT films with x-ray rocking curve widths of 72 arc seconds were observed. In general, the thin film dielectric constant at microwave frequencies is low (200-950) compared to the reported bulk value, but strongly dependent on the Sr/Ba ratio. Biasing of a ferroelectric interdigital capacitor (< 200 kV/cm) produces a change in the dielectric constant which resulted in a phase shift in the reflected signal (S_{11}) measured as a function of frequency from 100 MHz to 10 GHz. The dielectric loss tangent measurement, as measured at room temperature and 9.2 GHz, ranges from 0.1 to 1.2 x 10^{-3} and depends on the Sr/Ba ratio. These data show that SBT thin films are suitable for the development of frequency tunable microwave circuits and components.

INTRODUCTION

High quality ferroelectric thin films offer unique opportunities for the development of advanced microwave signal processing devices. In a ferroelectric the dielectric constant can be varied by applying an electric field. The variable dielectric constant results in a change in the phase velocity in the device allowing it to be tuned in real time for a particular application. The use of ferroelectric materials as a non-linear dielectric at microwave frequencies and the integration of tunable dielectrics with conductors that have low microwave surface resistance (R_S) is currently being investigated for a variety of advanced high frequency device applications [1-8]. Thin films offer a unique advantage over bulk materials for these applications. Large electric fields (0-200 kV/cm) can be achieved in thin films using much lower bias voltages (0-10 V). Small, compact, low power microwave devices that could be fabricated from structures based on ferroelectric films include phase shifters, tunable filters and tunable high Q resonators.

Several critical issues need to be addressed for the fabrication of ferroelectric thin films for microwave applications. These issues include the characterization of thin film properties at microwave frequencies to determine the dielectric constant, tunability, Curie temperature and dielectric losses. At high frequencies, conductors with low R_S are desirable. Ferroelectric materials are structurally compatible with high temperature superconductors which have demonstrated low surface resistance losses at microwave frequencies. Integration of ferroelectric thin films with high temperature superconductor materials can be implemented with bilayer structures for coplanar wave guides and trilayer structures for microstrip wave guides.

$Sr_xBa_{(1-x)}TiO_3$ (SBT) is ideally suited for the development of ferroelectric based microwave electronics. The Curie temperature of bulk SBT ranges from 30 to 400 K for x between 1 and 0, respectively. The ability to control the dielectric properties in a simple way will allow device structures to be easily optimized for maximum tunability and minimum loss at the desired frequency. We have investigated the growth of SBT thin films (x=0.2 - 0.8) by pulsed laser deposition (PLD) and characterized the dielectric constant, loss tangent and dc field effect at frequencies from 100 MHz to 10 GHz.

Mat. Res. Soc. Symp. Proc. Vol. 361 © 1995 Materials Research Society

EXPERIMENTAL

$Sr_xBa_{(1-x)}TiO_3$ films were deposited by PLD onto single crystal substrates of (100) MgO and (100) $LaAlO_3$ (LAO). The output of a short pulsed (30 ns FWHM) excimer laser operating on KrF (248 nm) was focused to an energy density of 1-2 J/cm^2 onto a stoichiometric target. SBT targets (x = 0.2 - 0.8) were prepared by mixing appropriate amounts of $SrTiO_3$ and $BaTiO_3$ powders, pressing 3/4" diameter targets to 15,000 pounds and then annealing overnight in flowing oxygen to a maximum temperature of 750 °C. The target substrate distance was fixed at 3 cm. Films were deposited in 350 mTorr of flowing oxygen (~ 10 sccm) onto heated substrates (750 - 850 °C) of MgO (100) and $LaAlO_3$ (100). Substrates were attached using silver print to a stainless steel block which was heated by the output from two 360 W quartz-halogen lamps. The laser repetition rate was 5-10 Hz. Film deposition rates were ~ 2.5 - 3 Å/laser pulse on 1.0-1.5 cm^2 substrates. X-ray diffraction patterns were obtained using Cu $K\alpha$ radiation from a rotating anode source using the standard θ– 2θ geometry.

High frequency measurements were made on both unpatterned SBT films and on patterned Au/SBT or SBT/YBCO bilayers . Loss tangent measurements were made on unpatterned films using a cavity perturbation technique at 9.2 GHz. Both the frequency shift and change in cavity Q were used to determine the real and imaginary parts of the susceptibility. For patterned structures, on top of the SBT films, a thick Au film was electrochemically deposited to a thickness of 1- 2 μm. Interdigital capacitors of various dimensions were patterned from the thick gold films and the reflection coefficient (S_{11}) was analyzed on an HP 8753A network analyzer from 100 MHz to 10 GHz.

RESULTS

SBT (x=0.2 - 0.8) films were deposited by PLD onto (100) MgO and $LaAlO_3$ substrates. The deposited films were from 3 to 5 μm thick. The deposited films were characterized by x-ray diffraction. Shown in Fig. 1 is the diffraction pattern for an SBT film deposited onto MgO. The film is single phase and epitaxial as shown by both the normal and in-plane alignment.

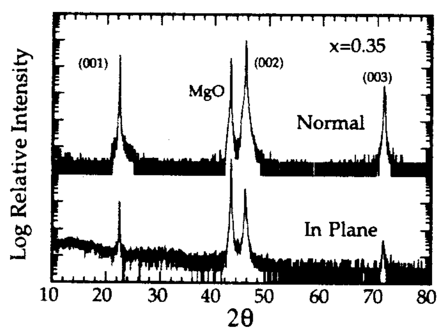

Figure 1. X-ray diffraction from an SBT (x=0.35) film deposited onto (100) MgO.

Typically, x-ray rocking curves for the SBT films ranged from 0.3 to 0.7° for the (002) reflection. However, for several films, the x-ray rocking curve was near or at the 1/6° resolution of the diffractometer. Further characterization of the film structure was made using monochromatic synchrotron radiation [9]. Figure 2 shows the rocking curve for the SBT (002) reflection measured with synchrotron radiation of 8.0 keV at the National Synchrotron Light Source using the X-14B beam line. The FWHM for SBT(002) was found to be 72 arc seconds and the substrate ($LaAlO_3$ (002)) to be 36 arc seconds. Although there is a large mismatch between the SBT film and the $LaAlO_3$ substrate (4%), the rocking curve width is comparable or better than that observed for semiconductor films grown by molecular beam epitaxy for a similar film-substrate mismatch and comparable film thickness.

In order to verify the monodomain character of the films, a double crystal topograph was obtained using an asymmetrically cut Si(111) crystal to monochromatize and expand the primary x-ray beam. The topograph was obtained using the SBT(002) reflection. Figure 3 shows the topograph for an $Sr_{0.5}Ba_{0.5}TiO_3$ film. Besides a few dislocations, the topograph reveals the single domain nature of the film.

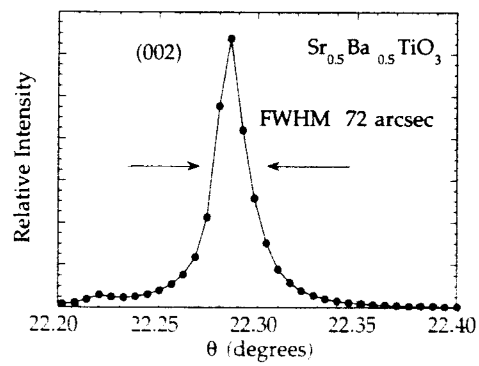

Figure 2. X-ray rocking curve for (002) reflection of SBT (x=0.5) film deposited onto LAO.

Figure 3 X-ray topograph (using (002) reflection) of SBT (x=0.5) film deposited onto LAO.

The lattice constants for the SBT (x=0.2 to 0.8) films were determined from the position of the (003) reflection. Shown in Fig. 4 is a plot of the variation in the lattice constant with composition for the deposited films in comparison with the lattice constant for the bulk material. A slight (0.3%) increase in the thin film lattice constant is observed over the range of compositions investigated. This variation may arise as a consequence of strain at the film substrate interface in the heteroepitaxial growth. As has been shown previously, the properties of the ferroelectric thin films are different from the bulk [3,8]. Reduced dielectric constants and a broader than bulk temperature dependence has been observed.

Capacitors fabricated from patterned electrochemically deposited Au capacitors grown on SBT films have been used to measure the electric field effect for SBT (x=0.2 - 0.8) films at room temperature (Fig. 5). A series of interdigital capacitors were fabricated in which the length and gap dimensions were varied from 5 - 15 μm and the length ranged from 10 to 150 μm. These structures produced a capacitance on the order of a few pf. The reflected signal (S_{11}) was measured as a function of frequency from 100 MHz to 10 GHz. The change in the phase of the reflected signal as a function of bias voltage is shown in Fig. 6. The change in phase is a result of the field-induced change in the dielectric constant of the ferroelectric thin film. For this particular structure, the maximum bias voltage of 10 V results in an electric field strength of 13 kV/cm. The change in capacitance for the 0 to 10 V tuning was calculated to be 2.45 to 2.19 pf or 11%.

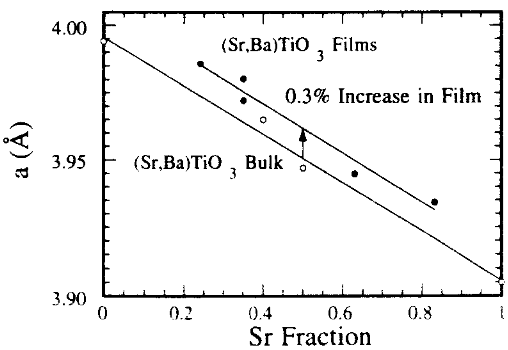

Figure 4. Variation of lattice constant for bulk SBT and thin films deposited onto (100) MgO and LAO substrates.

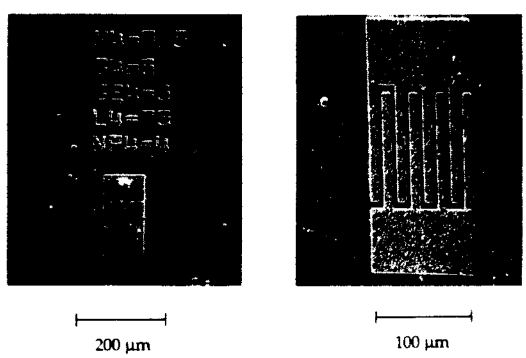

Figure 5. Thin film ferroelectric capacitors with thick (1.5 - 2.0 μm) electrodes.

Co-planar wave guide structures have also been fabricated for microwave band pass and band reject filters implemented with high temperature superconductors (HTS) as the conductor. On top of the patterned $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin film, ~0.5 μm thick SBT (x=0.1) thin film were deposited. Electrical contact to the YBCO film was made by ion milling through the ferroelectric. The filters had a centerband frequency at ~6 GHz and exhibited a 3% tuning range for a 0 - 50 V bias (0 - 100 kV/cm).

The loss tangent for SBT thin films was obtained at room temperature using a simple cavity perturbation technique. SBT films, (~ 3 - 5 μm thick) deposited onto MgO and LAO substrates, were cut into strips ~ 0.080" wide. Samples were introduced into a high Q TE_{103} cavity resonant at 9.3 GHz. Both the frequency shift and change in Q were used to calculate the sample susceptibility ($\varepsilon' + \varepsilon''$). The ratio of the real and imaginary parts was used to calculate the loss tangent ($\tan\delta=\varepsilon''/\varepsilon'$). For an MgO substrate, the cavity technique yielded a dielectric constant of 8.8 and a loss tangent of 2.2 x 10^{-5} which are comparable to values reported in the literature. The uncoated MgO substrate was replaced with an SBT (x=0.2 to 0.8) coated substrate. Film thickness were ~3 - 5 µm. The dependence of the dielectric constant and loss tangent on the films composition is shown in Fig. 7. The maximum dielectric constant measured (~950) is at x=0.2 and the loss tangent at this composition is 0.1. As the Sr fraction increases, both the loss tangent and the dielectric constant decrease to a minimum value of ~ 400 and 1.2 x 10^{-3} for the dielectric and loss tangent respectively. The general trends in the thin film properties are as expected from bulk behavior with a slight shift in the thin film Curie temperature. As can be seen from the plot, the maximum dielectric constant for the thin film occurs for x=0.2. Therefore, we can consider the Curie temperature for x=0.2 to be 298 K. As the Sr fraction is increased or decreased, the Curie temperature is shifted to higher or lower temperatures which is seen as a decrease in the dielectric constant measured at room temperature. For $x > 0.2$ the film is paraelectric at room temperature and for $x < 0.2$ the film is ferroelectric at room temperature. For microwave applications it has been proposed that the paraelectric phase is best as the absence of a remnant polarization may result in material with a lower loss tangent. Based on bulk data, low loss paraelectric films would be obtained at room temperature for $x \geq 0.4$.

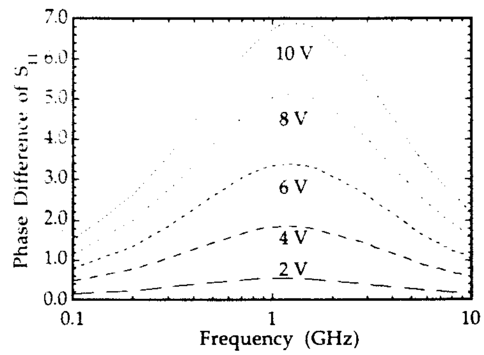

Figure 6. Change phase for reflected signal (S_{11}) for Au/SBT (x=0.35) capacitor as a function bias voltage.

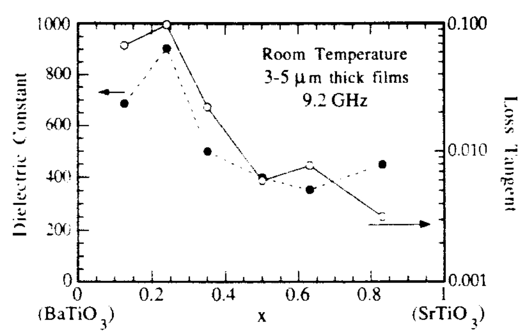

Figure 7. Variation of dielectric constant and loss tangent with composition for SBT films deposited onto MgO and LAO substrates measured at room temperature and 9.2 GHz.

CONCLUSION

High quality $Sr_xBa_{(1-x)}TiO_3$ thin films have been deposited by pulsed laser deposition onto single crystal substrates of (100) MgO and $LaAlO_3$. The deposited films are oriented both with respect to the substrate surface normal and the plane of the film. Highly oriented films have been deposited onto $LaAlO_3$ substrates and have been characterized by x-ray rocking curve widths for the (002) reflection of 72 arcsec. The deposited films exhibit a field dependent dielectric constant in coplanar microwave structures fabricated from both normal metal (Au) and HTS electrodes. The dielectric constant and loss tangent were evaluated at room temperature for values of x from 0.2 to 0.8. The maximum dielectric constant and loss tangent (1000 and 0.1 respectively) were observed for x=0.2. As the Sr fraction increased, both the dielectric constant and loss tangent decreased to approximately 400 and 1.2×10^{-3} respectively. The high frequency data show that ferroelectric thin films can be used to fabricate low loss active microwave devices.

ACKNOWLEDGMENTS

Support of this research has been provided by the Office of Naval Research and by Superconducting Core Technologies NCRADA-NRL-94-027.

REFERENCES

1. F. J. Elmer, S. J. Jang, Army Science Conference Proceedings, 283 (1988).
2. V. K. Varadan, D. K. Gohdgaonkar, V. V. Varadan, J. F. Kelly and P. Glikerdas, Microwave Journal, Jan. **116** (1992).
3. K. R. Carroll, J. M. Pond, D. B. Chrisey, J. S. Horwitz, R. E. Leuchtner, Appl. Phys. Lett., **62** 1845 (1993).
4. D. B. Chrisey, J. S. Horwitz, J. M. Pond, K. R. Carroll, P. Lubitz, K. S. Grabowski and R.E. Leuchtner, IEEE Trans. on Applied Superconductivity, **3** 1528 (1993).
5. C. M. Jackson, J. H. Kobayashi, A. Lee, C. Pettiette-Hall, J. F. Burch, R. Hū, R. Hilton, J. McDade, Microwave and Optical Tech. Lett. **5** 722 (1992).
6. D. Gait, J. C. Price, J. A. Beal, R. H. Ono, Appl. Phys. Lett., **63** 3078 (1993).
7. R. W. Babbit, T. E. Koscica, W. C. Drach, Microwave Journal, **35** 63 (1992).
8. J.S. Horwitz, D.B. Chrisey, J.M. Pond, R.C.Y. Auyeung, C.M. Cotell, K.S. Grabowski, P.C. Dorsey and M.S. Kluskens, Accepted for Publication, Integrated Ferroelectrics (1994).
9. S. B. Qadri, J.S. Horwitz, D.B. Chrisey, R.C.Y. Auyeung and K.S. Grabowski, Accepted for Publication, Appl. Phys. Lett. (1995).