

Microwave measurement of the dielectric constant of $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{TiO}_3$ ferroelectric thin films

K. R. Carroll, J. M. Pond, D. B. Chrisey, J. S. Horwitz, R. E. Leuchtner, and K. S. Grabowski
Naval Research Laboratory, Washington, DC 20375

(Received 7 December 1992; accepted for publication 9 February 1993)

Measurement of the relative dielectric constant of a $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{TiO}_3$ (SBT) thin film is presented as a function of electric field strength and temperature over a broad frequency range using a microstrip transmission line. The transmission line was fabricated from a trilayer structure where the SBT film, grown by pulsed laser deposition, was bounded by silver and platinum metallization layers. Such structures involving ferroelectric films could be useful for microwave applications because of the substantially smaller bias voltages ($\approx 1\text{--}10$ V) compared to those required for bulk material. The SBT film was found to exhibit a dielectric constant of $\approx 120\text{--}250$ and a large electric field modulation of $\approx 50\%$ at 200 kV/cm. These properties of the material as well as the Curie temperature are compared to those of bulk SBT.

Ferroelectrics are interesting for microwave devices because it is possible to change the dielectric constant by the application of an electric field. However, these materials have typically been of limited use in this area because of the large voltages, on the order of 1000 V, needed to bias bulk substrates in a microstrip geometry.¹ With thin films of these materials, the needed bias voltages are reduced to less than 10 V, which is compatible with the voltage requirements of present semiconductor-based systems. Among the more promising active microwave applications are electronically tunable phase shifters, mixers, delay lines, and filters. To help demonstrate the feasibility of employing ferroelectric thin films for frequency-agile microwave applications, the dielectric constant of a strontium barium titanate (SBT) thin film has been measured as a function of temperature and applied electric field. Another important issue for ferroelectric devices are the losses. The microstrip technique used here is not able to determine the dielectric losses due to the large attenuation of the signal by the silver Ag and platinum Pt metallization layers at these frequencies. While this demonstration structure provides a convenient method of characterizing the ferroelectric films, the integration of superconductors in these multilayer geometries will be required to achieve acceptable device performance.

For a transmission line in a microstrip configuration, the series impedance per unit length is given by^{2,3}

$$Z = j\omega\mu_0 g_1 + Z_m g_2 + Z_g/W \quad (1)$$

and the parallel admittance is given by

$$Y = \omega\epsilon_0(j\epsilon_{re} + \epsilon_r \tan \delta)/g_1, \quad (2)$$

where μ_0 and ϵ_0 are the permeability and permittivity of free space, respectively, ω is the angular frequency, ϵ_r the relative dielectric constant of the ferroelectric, ϵ_{re} an effective dielectric constant dependent on the geometry, and $\tan \delta$ the loss tangent of the ferroelectric. The factors g_1 and g_2 are dependent on the geometry and in the parallel plate limit, $g_1 = d/W$ and $g_2 = 1/W$, where d is the thickness of the dielectric and W is the width of the microstrip line. For our measurements, these geometric factors devi-

ated from the parallel plate values by less than 20%. In expression (1), Z_m and Z_g represent the surface impedance of the microstrip line and ground plane, respectively, which can be approximated by⁴

$$Z_s = \sqrt{\frac{j\omega\mu_0}{\sigma}} \coth(t\sqrt{j\omega\mu_0\sigma}), \quad (3)$$

where σ is the conductivity and t the thickness of the metal. The conductivity can be a complex number $\sigma = \sigma_1 - j\sigma_2$ which allows the above formalism to describe superconducting microstrip. For the transmission line to be considered below, we are in the thin-film limit where $Z_s \approx (\sigma t)^{-1}$.

From the above expressions, the characteristic line impedance $Z_L \equiv (Z/Y)^{1/2}$ and the complex phase $\Gamma \equiv (ZY)^{1/2}$ can be determined. Given these parameters for a transmission line connected to a 50- Ω system, the forward transmission coefficient S_{21} is⁵

$$S_{21} = \frac{4Z_L Z_0}{(Z_L + Z_0)^2 e^{\Gamma l} - (Z_L - Z_0)^2 e^{-\Gamma l}}, \quad (4)$$

where $Z_0 = 50 \Omega$ and l is the line length. By curve fitting the magnitude and phase of S_{21} , the SBT dielectric constant and effective conductivity of the metals can be determined.

For the microstrip line, the platinum ground plane and the SBT layer were deposited by pulsed laser deposition (PLD)⁶ using a KrF excimer laser. PLD is a versatile film-growth technique for multicomponent oxides as has been demonstrated by the deposition of other ferroelectrics, ferrites, and the high temperature superconductors.⁶⁻⁸ The growth condition for the platinum layer was 450 °C in 50 mTorr of argon onto a (100) MgO substrate.⁹ These conditions resulted in a (100) Pt film with a 300-nm thickness. The SBT dielectric was then ablated from a stoichiometric $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{TiO}_3$ target with a substrate temperature of 750 °C in 300 mTorr of oxygen for which the resulting film thickness was 400 nm. During this part of the processing, a shadow mask was used to prevent the SBT from covering the entire platinum layer and conse-

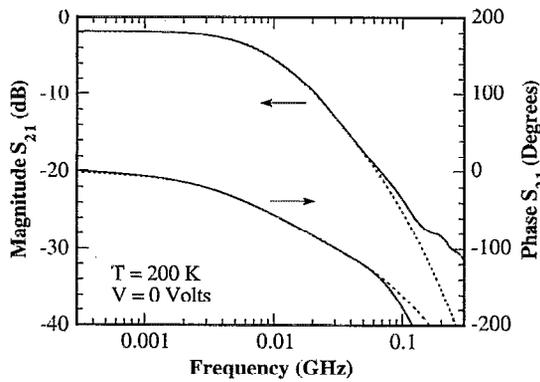


FIG. 1. Magnitude and phase of the transmission coefficient S_{21} with no applied bias at 200 K.

quently allowed direct electrical connection to the ground plane. A microstrip line with a width of $10.8 \mu\text{m}$, a length of 1.00 cm, and a thickness of 750 nm was defined by a two-level-photoresist lift-off process from an e -beam evaporated Ag film.

Gold bond wires with a $25\text{-}\mu\text{m}$ diameter provided the electrical connection between the microstrip line and the rf connectors of the test fixture. The test fixture was thermally attached to a closed cycle refrigerator which allowed the sample temperature to be controlled above and below the Curie temperature of the SBT film. To make the transmission measurements, a vector network analyzer was used where the cables and connectors leading to the test fixture were calibrated out of the measurement. The measurements were independent of the incident rf power for levels below 10 mW. The dc bias was applied through a bias tee connected to the rf cables.

In Fig. 1, the measured amplitude and phase of the transmission characteristics S_{21} is shown when there is no bias voltage at a temperature of 200 K. The dashed lines indicate the results predicted by Eq. (4). These curves were obtained with a relative dielectric constant of 250. As can be seen from the graph, there is excellent agreement between the model and the measured data below ≈ 0.07 GHz. These results imply that the dielectric constant is frequency independent between 0.3 and 70 MHz. Since the attenuation of the transmission-line mode grows rapidly with frequency, the deviation from the model for higher frequencies is thought to result from transmission through a parallel path. This parallel path had a resonant frequency at ≈ 1 GHz.⁸ For bulk SBT materials, the permittivity has been found to be constant for frequencies above 10 GHz.¹ The effective conductivity for the microstrip line can also be determined from the transmission measurements. Values for the metal film conductivities were within a factor of two of bulk values.

With the use of Eq. (4), the dielectric constant vs temperature can be calculated and is shown in Fig. 2 for several bias voltages. The Curie temperature T_c is near 200 K and the maximum dielectric constant is 250 at zero field strength and 200 K. One notes that the peak in the dielectric constant is quite broad and does not show the $(T - T_c)^{-1}$ behavior expected for a ferroelectric. In spite of

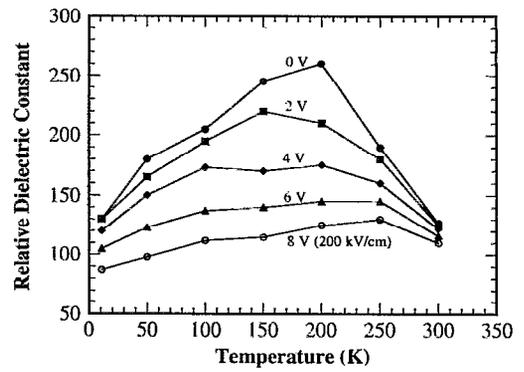


FIG. 2. Relative dielectric constant of the $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{TiO}_3$ film as a function of temperature for several applied fields.

these differences, the electric field modulation of the dielectric constant is as large as 50%. For comparison, bulk SBT material with the same composition exhibits dielectric constants on the order of 1000 near the Curie temperature of ≈ 275 K.¹ It is also interesting to note that the maximum applied field of 200 kV/cm is a factor of 10 greater than that needed to induce a similar percentage change in the dielectric constant of bulk SBT. Stress and compositional inhomogeneities in the film may account for some of the deviations from the bulk properties. These factors can result in a variation of the Curie temperature throughout the film preventing the Curie-Weiss dependence from being observed. Another possibility is that a space charge layer has formed between the normal metals and the SBT film. This layer suppresses the measured dielectric constant by screening the applied field from the interior of the film. Although this mechanism might account for a different ϵ_r , it would not account for the shift in T_c and the broadening of the Fig. 2 curves.

In Fig. 3, the inverse of the dielectric constant is plotted as a function of the applied field. This data can be fit to a second-order polynomial in the field. For temperatures above the Curie temperature (200, 250, and 300 K) the field dependence is almost linear, and for temperatures below the transition temperature, there is a definite parabolic trend to the data. From the lowest order field expansion of the free energy, one expects the opposite behavior to occur,

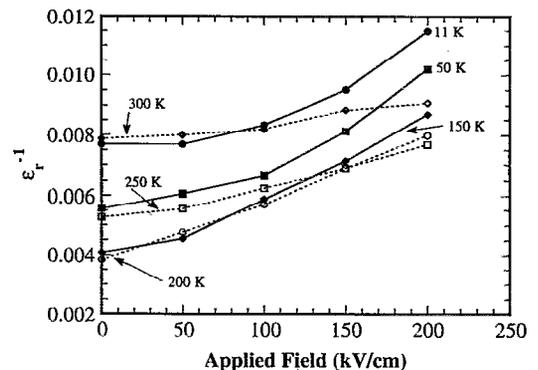


FIG. 3. Reciprocal of the relative dielectric constant as a function of field strength for several temperatures.

i.e., a linear field dependence below the transition temperature and a parabolic dependence above for $1/\epsilon_r$.¹⁰

The hysteresis in the dielectric constant as a function of voltage was also investigated. A self bias of ≈ 20 kV/cm was found at 11 K and gradually vanished as the Curie temperature was approached. While the polarization vs applied field was far from an ideal square loop, the coercive field could be determined from the maxima in the dielectric constant. The value was ≈ 60 kV/cm at 11 K. A determination of the spontaneous polarization was prevented by the nonideal nature of the hysteresis curve.

As has been noted in the above paragraphs, the properties of thin-film SBT differ substantially from its bulk counterpart. Obviously, additional film characterization will be needed before the intrinsic properties of the thin films can be completely understood. However, a few of these differences are advantageous for microwave applications. The lower dielectric constant allows easier impedance matching to 50- Ω systems and the broader temperature response shown in Fig. 1 will make any device design less sensitive to a variation in Curie temperature induced by compositional differences or defects. Also, the amount of temperature control required for stable device operation will be substantially less in the thin-film case.

While thin ferroelectric films allow one to employ reasonable bias voltages, the losses in normal conductors preclude the use of such a structure in the microwave frequency range. As can be seen from the magnitude of S_{21} , one has 30 dB of attenuation for a relatively low frequency of 100 MHz and a short 1-cm line length. These losses indicate the need for a superconductor to replace the normal metals in these structures. For this case, the analysis that has been done for superconducting parallel-plate transmission line is directly applicable.¹¹ This analysis indicates that the superconductor losses are much less than the ferroelectric losses. If the thin-film loss mechanism is similar to that of bulk SBT material, the transmission line performance would then be limited by a loss tangent less than 0.02.¹ For many applications, this amount of attenuation would be acceptable. The use of superconductors also places a constraint on the Curie temperature of the ferroelectric. To achieve the maximum voltage modulation of the dielectric constant with the high temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$, e.g., one would want the Curie temperature between ≈ 77 and 90 K. Given that the Curie temperature of BaTiO_3 is ≈ 400 K¹⁰ and SrTiO_3 is ≈ 35 K,¹² the T_c of a SBT film can be lowered to this tempera-

ture range by adjusting the strontium to barium ratio.

In conclusion, the 50% voltage modulation of the relative permittivity is very promising for many microwave applications providing that the ferroelectric films can be integrated with superconducting films. Currently, the compatibility of SBT films with the high temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$ is being investigated. Since both $\text{YBa}_2\text{Cu}_3\text{O}_7$ and SBT have perovskite structures, the materials are expected to grow epitaxially as the trilayer is formed. With the use of superconducting films for the metalization layers, one would be able to determine any frequency dependence of the dielectric constant above 0.1 GHz and the losses of the SBT film. Additional questions concerning the dependence of the Curie temperature and other film parameters on composition and film defects also require investigation.

The support of the Office of Naval Research is gratefully acknowledged and several useful discussions with Dr. Mike Bell about the properties of ferroelectrics are also greatly appreciated. This work was done while one of the authors (K.R.C.) held a Naval Research Laboratory/National Research Council research associateship.

- ¹V. K. Varadan, D. K. Ghodgaonkar, V. V. Varadan, J. F. Kelly, and P. Glikerdas, *Microwave J.* **35**, 116 (1992); R. W. Babbit, T. E. Kosciwa, and W. C. Drach, *ibid.* **35**, 63 (1992).
- ²K. C. Gupta, R. Garg, and I. J. Bahl, *Microstrip Lines and Slotlines* (Artech House, Dedham MA, 1979).
- ³R. A. Pucel, D. J. Masse, and C. P. Hartwig, *IEEE Trans. Microwave Theory Tech.* **16**, 342 (1968).
- ⁴R. E. Matick, *Transmission Lines for Digital and Communication Networks* (McGraw-Hill, New York, 1969), p. 243.
- ⁵R. E. Collin, *Foundations for Microwave Engineering* (McGraw-Hill, New York, 1966).
- ⁶D. B. Chrisey, J. S. Horwitz, H. S. Newman, M. E. Reeves, B. D. Weaver, K. S. Grabowski, and G. P. Summers, *J. Supercond.* **4**, 57 (1991).
- ⁷J. S. Horwitz, K. S. Grabowski, D. B. Chrisey, and R. E. Leuchtner, *Appl. Phys. Lett.* **59**, 1565 (1991); R. Ramesh, K. Luther, B. Wilkins, D. L. Hart, E. Wang, J. M. Tarascon, A. Inam, X. D. Wu, and T. Venkatesan, *ibid.* **57**, 1505 (1990).
- ⁸D. B. Chrisey, J. S. Horwitz, J. M. Pond, K. R. Carroll, P. Lübitz, K. S. Grabowski, R. E. Leuchtner, C. A. Carosella, and C. V. Vittoria, *IEEE Trans Appl. Supercond.* **3**, 1528 (1993).
- ⁹R. E. Leuchtner, D. B. Chrisey, J. S. Horwitz, and K. S. Grabowski, *Surf. Coatings Technol.* **51**, 476 (1992).
- ¹⁰M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials* (Oxford University Press, Oxford, 1977).
- ¹¹K. R. Carroll, J. M. Pond, and E. J. Cukauskas, *IEEE Trans. Appl. Supercond.* **3**, 9 (1993); J. M. Pond, J. H. Claassen, and W. L. Carter, *IEEE Trans. Microwave Theory Tech.* **35**, 1256 (1987).
- ¹²H. E. Weaver, *J. Phys. Chem. Solids* **11**, 274 (1959).