

High tunability barium strontium titanate thin films for rf circuit applications

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Large variations in the permittivity of rf magnetron sputtered thin-film barium strontium titanate have been obtained through optimization of growth conditions for maximum dielectric strength and zero-field permittivity in a parallel-plate capacitor structure. Using nominal target compositions of $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$, and Pt electrodes on *c*-plane sapphire substrates, adjustment of the O_2 partial pressure during deposition was used to vary the excess Ti incorporation into the films, which influenced the low-field permittivity, loss tangent, and dielectric strength. By balancing the benefits of a high permittivity with dielectric strength and loss, we have produced films capable of sustaining short-duration fields greater than 4 MV/cm with over 13:1 (>90%) change in dielectric constant, and greater than 5:1 tunability in bias fields under 1 MV/cm. © 2004 American Institute of Physics. [DOI: 10.1063/1.1818724]

Barium strontium titanate (BST) is a solid solution perovskite with a field-dependent permittivity. In recent years, there has been a strong interest in thin-film BST, first for dynamic random access memory¹ and later rf or microwave^{2,3} applications. While much work has focused on reducing material losses in thin films for microwave devices,⁴ the tunability (change in dielectric constant with bias) is also a critical parameter in determining overall circuit loss in an application. Figures of merit involving products of intrinsic material quality and tunability are sometimes proposed for comparing materials, but these are not always useful predictors of circuit performance. At high frequencies (in the microwave range), ohmic losses in the capacitor electrodes become a significant loss contributor, mitigating the advantages of low loss-tangent films. Higher tunabilities allow for designs with fewer cascaded tuning elements, directly reducing the net circuit loss. Phase shifter circuits, for example, require a predetermined amount of phase delay, and a number of individual phase shifting units are cascaded to achieve this goal. When the film tunability is increased, the amount of phase shift per unit is increased, thereby decreasing the total number of units required.⁵

Sputtered BST films have been investigated by several researchers.^{4,6–8} The role of the O_2 ambient during growth has been specifically studied for its influence on leakage,⁹ lifetime,¹⁰ and film texturing.¹¹ In this letter, the electrical tunability of BST capacitors with Pt electrodes was optimized with respect to O_2 partial pressure during sputter deposition, leading to significantly higher tunabilities than previously reported. The effects of O_2 concentration during sputtering appear to be complex; the ambient O_2 partial pressure influences not only the oxygen incorporation into the films, but also the Ti nonstoichiometry and the *A/B* site ratio which are strongly linked to the zero-field permittivity and loss.¹² Additionally, evidence of changes in the Pt electrode morphology when exposed to O_2 at high temperatures has been reported;¹³ thus, the resulting film texture and interface quality are influenced by the O_2 partial pressure.

The BST films used in this work were deposited by rf magnetron sputtering. The films were simultaneously co-sputtered from two 3 in. ceramic targets on to both platinized and bare *c*-plane sapphire substrates in an Ar/ O_2 ambient. The two targets had slightly different compositions, one being $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ (stoichiometric) and the other $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Ti}_{1.02}\text{O}_3$ (2% excess Ti). The targets were located 30° off axis and the target-to-substrate distance was 4.25 in. The Ar and O_2 flow rates were adjusted for a total pressure of 45 mTorr for all growths in the series; the Ar/ O_2 flow rates for the films in this set were 90/10, 80/20, 70/30, 60/40, and 50/50 sccm corresponding to O_2 partial pressures of 4.5, 9.0, 13.5, 18.0, and 22.5 mTorr, respectively. The substrate temperature (700 °C), rf power (150 W on each gun), and growth time (64 min) were held constant. The resulting film thicknesses were determined by surface profilometry, and ranged from 113 to 145 nm with lower O_2 partial pressures corresponding to higher deposition rates.

Parallel-plate capacitors were fabricated using the samples grown on platinized sapphire in a two-layer mask process. The first layer defined BST mesas for a device isolation etch in buffered HF. The second mask layer opened windows for lift-off of mesa and ground plane electrodes. 150 nm thick Pt electrodes were deposited by electron-beam evaporation. No specific processing steps were taken to improve the electrode–film interface properties.

Capacitance and *Q*-factor (inverse loss tangent) tuning curves were measured using an Agilent 4294A impedance analyzer for low-voltage measurements at 1 MHz and 100 MHz with a 500 mV oscillation amplitude. High-voltage (>40 V) measurements at 100 MHz were obtained using an HP 8722D network analyzer with an external bias tee. The high voltages were sourced using a Keithley 6517A electrometer with a series resistance of approximately 50 kΩ. Measurements of dc leakage currents were performed using an Agilent 4155B semiconductor parameter analyzer. Compositional data were determined through Rutherford back-scattering spectroscopy (RBS) analysis on the samples grown on bare sapphire.

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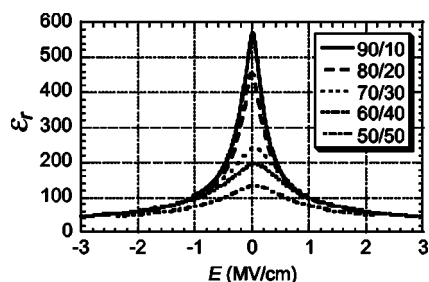
TABLE I. Compositional and electrical film properties for five films grown at the same total pressure of 45 mTorr, but with different Ar/O₂ flow rates.

Ar/O ₂ (sccm)	P _{O₂} (mTorr)	Ba/ Sr	(Ba+Sr)/ Ti	ϵ_r	Q (1 MHz)	Tunability (100 MHz)
90/10	4.5	0.94	1.00	571	41	11.36:1(91.2%)
80/20	9.0	0.93	0.93	455	75	13.71:1(92.7%)
70/30	13.5	0.92	0.88	241	161	7.88:1(87.3%)
60/40	18.0	0.89	0.82	194	159	4.90:1(79.6%)
50/50	22.5	0.87	0.78	134	42	3.74:1(73.3%)

Table I summarizes the electrical measurement and RBS analysis results. Electrical data in each row were verified on multiple devices from each sample. In materials such as BST with the ABO₃ perovskite structure, the ratio of A to B site ions is an important predictor of the defect density and quality of the film structure.¹⁴ RBS sensitivity to oxygen is poor, making the most useful results of the analysis the A-to-B site ratio, and in the case of a solid solution such as BST, the ratio of the different cations occupying the A site as well.

As the O₂ partial pressure was increased from 4.5 mTorr to 22.5 mTorr, the Ti nonstoichiometry increased, as indicated by the decrease in the A-to-B site ratio from unity to 0.78. Figures 1 and 2 show the effect of the excess Ti on the electrical properties of the film. The ϵ_r -E curves in Fig. 3 were obtained by progressively increasing the voltage sweep until catastrophic device failure. The tunability figures reported in Table I were then determined by taking the ratio of the maximum (zero-bias) permittivity to the minimum permittivity at breakdown. This follows the convention in the literature, but it is not a practical method for characterizing the “useful” portion of the ϵ_r -E curve. Figure 2 gives some insight in this regard; device Q-factor decreases dramatically above a certain critical field, which is associated with an exponential increase in leakage currents. Determination of the maximum sustainable field in the films for a practical application would require careful long-duration stress testing, but a rough approximation would be the field at which the leakage losses approach the intrinsic dielectric loss. For the best films in our series, this happens around 1 MV/cm.

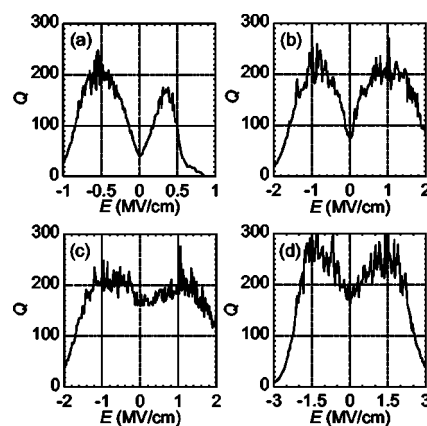
Maximum tunability measurements depicted in Fig. 3 show that despite its more desirable zero-bias dielectric properties, the 90/10 film ultimately had a lower tunability (11.6:1 or 91%) than the 80/20 film (13.7:1 or 93%) due to its lower dielectric strength. All of the films in the series asymptotically approach the same high-field permittivity limit, as shown in Fig. 1, but the 90/10 film broke down before approaching this limit. Figure 2 indicates the onset of

FIG. 1. 1 MHz tuning curves for films grown with different Ar/O₂ flow rates (sccm).

significant leakage in the 90/10 film at lower fields, so its “useful” tuning range is smaller as well. Although additional improvement in the breakdown voltage and leakage resistance was observed with the further incorporation of excess Ti into the film, the significant decrease in the zero-field permittivity limits the usefulness of such an approach for high tunability applications. Thus, the 80/20 film appears to be an optimum combination of tunability and dielectric strength for this series.

The large tunabilities in Table I exceed other reported data,^{15,16} due to the simultaneous improvement in both zero-bias permittivity and dielectric strength. In addition to stoichiometry, interface quality is thought to play a strong role in these results. The so-called “dead-layer” effect—a nontunable interfacial capacitance—appears to be influenced by interfacial roughness and surface contamination, and this in turn influences the apparent zero-bias dielectric constant.^{6,17} Interface quality also influences the effective Schottky barrier at the contact, which determines the amount of charge injection at high fields. Thus the maximum sustainable voltage and, therefore, tunability is also influenced by interface quality.

Figure 4 shows the transient leakage behavior⁸ of the films at a constant electric field of 200 kV/cm. Three different types of transient behavior were observed, with the most stoichiometric film having the sort of long transient we have previously reported.¹⁸ Note that there was no further decrease in leakage at this field for O₂ partial pressures above 13.5 mTorr (Ar/O₂ 70/30), with the measurements on the films with the three highest O₂ partial pressures being essentially indistinguishable. The initial benefit was quite dramatic, with an order of magnitude difference in leakage be-

FIG. 2. 1 MHz Q-factors for four of the films from Fig. 1: (a) Ar/O₂ 90/10 sccm, (b) Ar/O₂ 80/20 sccm, (c) Ar/O₂ 70/30 sccm, and (d) Ar/O₂ 60/40 sccm.

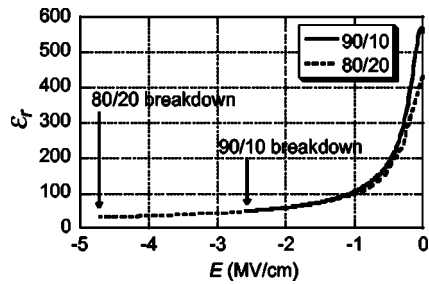


FIG. 3. 100 MHz maximum tuning curves for two most stoichiometric films, Ar/O₂ 90/10 and 80/20 sccm.

tween the two films with the lowest O₂ partial pressures. One possible explanation for this observed improvement is that small quantities of excess Ti can be accommodated in the grain boundaries, making them more insulating, but large amounts of excess Ti result in accommodation within the grain interiors.¹⁹ Investigation of the impact of the excess Ti on the film microstructure and crystallinity is currently underway.

In summary, we have demonstrated high tunability BST films deposited by rf-magnetron sputtering. Through variation of the O₂ partial pressure, we have shown that growth conditions can be optimized to yield very high tunability films with moderate loss, or low loss films with moderate tunability. A slight increase in the O₂ partial pressure beyond that required for a stoichiometric film (90/10) resulted in a film (80/20) with lower loss and lower leakage. This film exhibited a modest reduction in permittivity which was compensated for by an increase in dielectric strength, ultimately allowing for an extremely large tunability of 13.7:1. The films grown at higher O₂ partial pressures had increasing

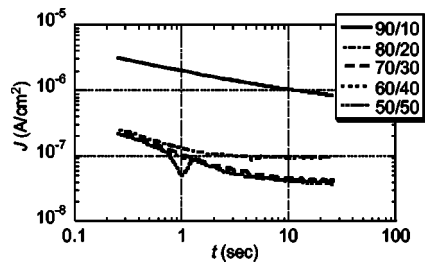


FIG. 4. Transient dc leakage current density at a constant field of 200 kV/cm for films grown with different Ar/O₂ flow rates (sccm).

excess Ti incorporation, resulting in lower tunabilities but higher *Q*-factors. For applications such as phase shifters, the benefits of high tunability dominate over the drawbacks of increased film loss, making our films well suited to such applications.

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- ¹D. E. Kotecki, J. D. Baniecki, H. Shen, R. B. Laibowitz, K. L. Saenger, J. J. Lian, T. M. Shaw, S. D. Athavale, C. Cabral, Jr., P. R. Duncombe, M. Gutsche, G. Kunkel, Y. J. Park, Y. Y. Wang, and R. Wise, *IBM J. Res. Dev.* **43**, 367 (1999).
- ²B. Acikel, T. R. Taylor, P. J. Hansen, J. S. Speck, and R. A. York, *IEEE Microw. Wirel. Compon. Lett.* **12**, 237 (2002).
- ³S. W. Kirchoefer, E. J. Cukauskas, N. S. Barker, H. S. Newman, and W. Chang, *Appl. Phys. Lett.* **80**, 1255 (2002).
- ⁴J. Im, O. Auciello, and S. K. Streiffer, *Thin Solid Films* **413**, 243 (2002).
- ⁵A. S. Nagra and R. A. York, *IEEE Trans. Microwave Theory Tech.* **49**, 1705 (1999).
- ⁶J. C. Shin, J. Park, C. S. Hwang, and H. J. Kim, *J. Appl. Phys.* **86**, 506 (1999).
- ⁷S. Zafar, B. Hradsky, D. Gentile, P. Chu, R. E. Jones, and S. Gillespie, *J. Appl. Phys.* **86**, 3890 (1999).
- ⁸M. C. Werner, I. Bannerjee, R. Zhang, P. C. McIntyre, N. Tani, and M. Tanimura, *J. Appl. Phys.* **89**, 2309 (2001).
- ⁹J. Lee, Y. C. Choi, and B. S. Lee, *Jpn. J. Appl. Phys., Part 1* **36**, 3644 (1997).
- ¹⁰M. S. Tsai, S. C. Sun, and T. Y. Tseng, *J. Appl. Phys.* **82**, 3482 (1997).
- ¹¹P. Padmini, T. R. Taylor, M. J. Lefevre, A. S. Nagra, R. A. York, and J. S. Speck, *Appl. Phys. Lett.* **75**, 3186 (1999).
- ¹²T. R. Taylor, P. J. Hansen, N. Pervez, B. Acikel, R. A. York, and J. S. Speck, *J. Appl. Phys.* **94**, 3390 (2003).
- ¹³N. Sugii and K. Takagi, *Thin Solid Films* **323**, 63 (1998).
- ¹⁴A. J. Moulson and J. M. Herbert, *Electroceramics* (Chapman and Hall, New York, 1990).
- ¹⁵A. Tombak, J. P. Maria, F. Ayguavives, J. Zhang, G. T. Stauff, A. I. Kingon, and A. Mortzawi, *IEEE Microw. Wirel. Compon. Lett.* **12**, 3 (2002).
- ¹⁶J. Im, O. Auciello, P. K. Bauman, S. K. Streiffer, D. Y. Kaufman, and A. R. Krauss, *Appl. Phys. Lett.* **76**, 625 (2000).
- ¹⁷S. K. Streiffer, C. Basceri, C. B. Parker, S. E. Lash, and A. I. Kingon, *J. Appl. Phys.* **86**, 4565 (1999).
- ¹⁸N. K. Pervez, P. J. Hansen, T. R. Taylor, J. S. Speck, and R. A. York, *Integr. Ferroelectr.* **53**, 503 (2003).
- ¹⁹S. Stemmer, S. K. Streiffer, N. D. Browning, and A. I. Kingon, *Appl. Phys. Lett.* **74**, 2432 (1999).