

Effect of SrTiO_3 Deposition Temperature on the Dielectric Properties of $\text{SrTiO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{LaAlO}_3$ Structures

F.A. Miranda
*Lewis Research Center
Cleveland, Ohio*

C.H. Mueller, R.E. Treece, T.V. Rivkin, and J.B. Thompson
*Superconducting Core Technologies
Golden, Colorado*

H.R. Moutinho
*National Renewable Energy Laboratory
Golden, Colorado*

*M. Dalberth and C.T. Rogers
University of Colorado
Boulder, Colorado*

Prepared for the
Eighth International Symposium on Integrated Ferroelectrics
sponsored by the University of Colorado
Tempe, Arizona, March 17–20, 1996



EFFECT OF SrTiO₃ DEPOSITION TEMPERATURE ON THE DIELECTRIC PROPERTIES OF SrTiO₃/YBa₂Cu₃O_{7-δ}/LaAlO₃ STRUCTURES

F. A. MIRANDA

National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135

C. H. MUELLER, R. E. TREECE, T. V. RIVKIN, and J. B. THOMPSON

Superconducting Core Technologies
720 Corporate Circle
Golden, Colorado 80401

H. R. MOUTINHO

National Renewable Energy Laboratory
1617 Cole Boulevard
Golden, Colorado 80401

M. DALBERTH and C. T. ROGERS

University of Colorado
Boulder, Colorado 80309

SUMMARY

We report on the effect of the deposition temperature of SrTiO₃ on the dielectric properties of SrTiO₃/YBa₂Cu₃O_{7-δ}/LaAlO₃ thin film multilayer structures. In these structures, the YBa₂Cu₃O_{7-δ} (YBCO) films were deposited at 800°C by laser ablation, followed by the in-situ deposition of the SrTiO₃ (STO) layer at one of the following temperatures: 750°C, 650°C, 550°C, 450°C, 350°C, and 250°C. Gold (Au) films were deposited and patterned on top of the STO layer to form planar Au/STO/YBCO capacitor structures. The electrical response was studied by measuring the dielectric constant (ϵ_r) and loss tangent ($\tan\delta$) of the ferroelectric film from 300-40 K, at 1.0 MHz, and at electric fields up to 100 kV/cm. Our results show 750°C to be a deposition temperature which allows for large variations of ϵ_r with limited enhancement of $\tan\delta$, while lower deposition temperatures cause a reduction of the induced change in ϵ_r and an increase in $\tan\delta$ with applied field.

INTRODUCTION

The study of the properties of ferroelectric thin films has generated enormous interest because of their potential use in areas such as phase shifters, ferroelectric capacitors, and memory devices, amongst others¹⁻³. The successful insertion of this technology in current systems demands full optimization of the material and electrical properties of the ferroelectric films. Of paramount importance for application of this technology in tunable microwave

components is the attainability of in-plane epitaxy with the host substrate or oxide layer, since it favors the growth of a ferroelectric film with a highly textured structure.^{4,5} Compatibility with other materials whose properties could also result in the optimization of the working system is also critical. High Temperature Superconductors (HTS) exhibit properties that could advantageously complement those of the ferroelectric films to enhance the functionality of electronic components currently in use. The superior performance of microwave components fabricated using HTS as compared to that of their conventional conductor counterparts, has been already demonstrated^{6,7}. Therefore, fabrication and characterization of HTS/ferroelectric thin film structures for the development of highly reliable, low loss, tunable microwave components, have become an area of intense research⁸⁻¹⁰. Because of a close lattice match ($\sim 2\%$)¹¹, as well as chemical compatibility between SrTiO₃ (STO) and the YBa₂Cu₃O_{7- δ} (YBCO) HTS thin films, these two materials could be favorably used to fabricate tunable microwave devices. Consequently, knowledge of the optimal deposition conditions for the growth of high quality STO films on YBCO thin films is of foremost importance.

In this paper, we present the results of our study on the effect of the STO deposition temperature on the electrical properties of SrTiO₃/YBa₂Cu₃O_{7- δ} /LaAlO₃ (STO/YBCO/LAO) thin film multilayer structures. Our results indicate that to obtain high quality STO films which allow for a large variation of ϵ_r with limited enhancement in $\tan\delta$ while preserving the integrity of the underlying superconducting film, the STO films should be deposited at temperatures near 750°C. Details on the film deposition process and multilayer structural characterization will be presented.

EXPERIMENTAL

All the STO/YBCO/LAO structures investigated in this study were fabricated in-situ by laser ablation. The structure was formed by growing the YBCO films on 4.0 mm x 10 mm x 0.50 mm (100) LaAlO₃ single-crystal substrates, which were held at temperatures of 800°C, followed up by the corresponding STO film. The same procedures were followed for the fabrication of all the samples except for the deposition temperature of the STO films. This was different for every structure, ranging from 750°C down to 250°C at intervals of 100°C. Typically, thicknesses of 350 nm and 500 nm were obtained for the YBCO films and the STO films, respectively.

The crystal structure and the surface topology of the STO films were analyzed by X-Ray Diffraction (XRD) and Atomic Force Microscopy (AFM), respectively. For the electrical characterization, thirty 400 μ m x 400 μ m x 2.5 μ m gold (Au) contacts were deposited by electron beam evaporation onto the STO layer. An area of approximately 1.0 mm x 4.0 mm of the STO films was chemically etched using a 7% solution of hydrofluoric acid (HF) to expose the underlying YBCO upon which a 2.5 μ m Au counterelectrode was subsequently evaporated (see Figure 1). This configuration was used to measure the relative dielectric constant (ϵ_r) and the loss tangent ($\tan\delta$) of the STO layer as a function of an externally applied dc electric field (\mathbf{E}), at 1.0 MHz, and in the temperature range from 300 to 40 K. During the measurements, the Au electrode was kept at a positive bias with respect to the YBCO electrode. The electrical measurements were performed using an HP-4192 A LF impedance analyser and a closed-cycle helium gas refrigerator to allow for a continuous

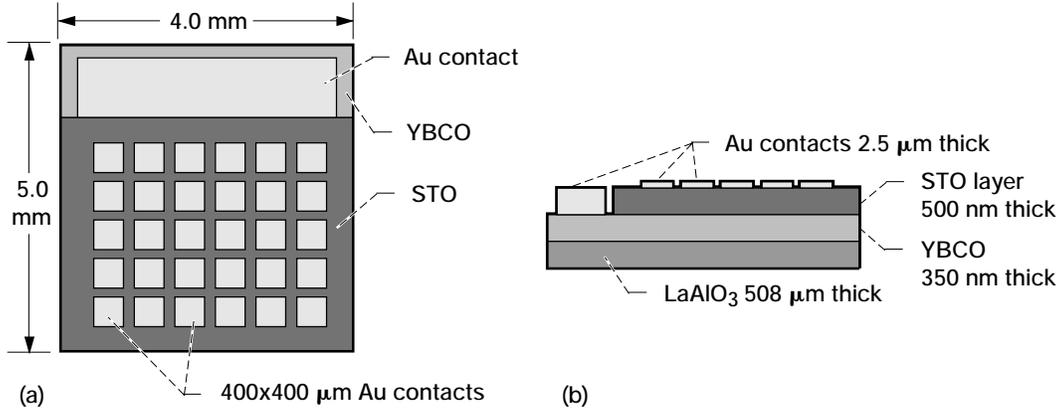


Figure 1.—Schematic representation of the configuration used to measure the electrical response of the STO layer in the STO/YBCO/LAO multilayer structure: (a) top view (b) side view.

temperature sweep within the aforementioned temperature range. For the measurements, the sample was mounted on a brass sample holder which in turn was bolted to the cold finger of the refrigerator. Electrical feedthroughs at the edges of the holder allowed for electrical contact to the Au dots across the surface of the STO sample. The measurement process was fully automated and controlled by an HP 9000-300 computer. Data were taken both during the cooling and thermal cycles to account for thermal hysteresis.

To examine the integrity of the underlying YBCO thin film after the deposition of the STO film, a section of the remaining multilayer film was used to measure the transition temperature (T_c) of the YBCO film by etching part of the top STO layer as explained above. Electrical contact was achieved by hot pressing 2 mil diameter gold wire to the YBCO film. These measurements were performed using a standard four-point probe technique.

RESULTS

The values of ϵ_r and $\tan\delta$ for the STO films, as well as their induced variation upon the application of a dc electric field, were highly dependent on the STO growth temperature. Figure 2 shows ϵ_r versus temperature for different electric field intensities applied across the STO film deposited at 750°C. Thermal hysteresis was not significant as evidenced by the good agreement between the data taken during the cooling and warming cycles for the field range shown in Figure 2. The ϵ_r of the STO film decreased by increasing the electric field, and the temperature at which the maximum ϵ_r was observed shifted to higher temperatures with increasing electric field. At 60 K, the change in intensity of the dc electric field decreased ϵ_r from 222 to 146. The $\tan\delta$ corresponding to this film is shown in Figure 3. These data show that $\tan\delta$ increased with respect to the zero field values by increasing the intensity of the electric field up to 8×10^4 V/cm. However, further increases in bias caused $\tan\delta$ to decrease slightly. At 60 K, $\tan\delta$ ranged from 0.015 to 0.123 depending on bias. The origin of the high values of $\tan\delta$, as compared to those observed in bulk STO, will be addressed in the discussion part of this paper. AFM images of the STO layer prior to the Au deposition (Figure 4) showed that the root mean square (rms) surface roughness of the STO film was 32 nm, and outgrowths up to 140 nm were apparent. XRD measurements show that this

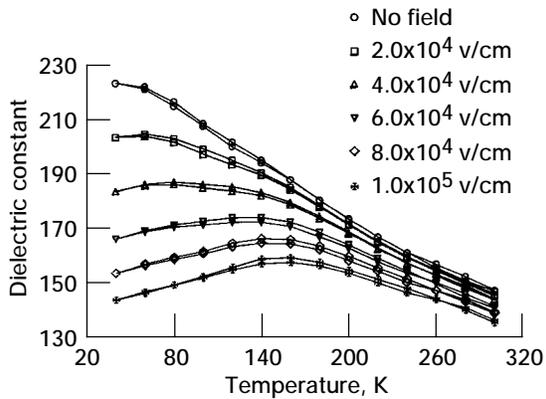


Figure 2.—Relative dielectric constant as a function of temperature and electric field for a 750 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

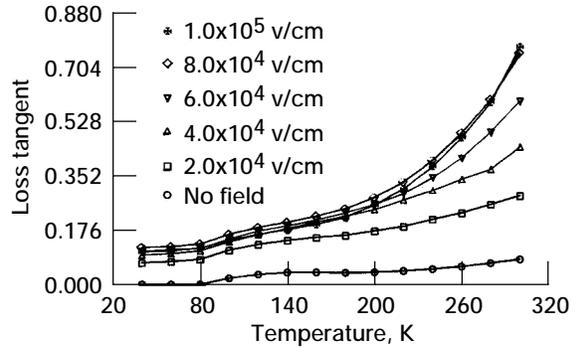


Figure 3.—Loss tangent as a function of temperature and electric field for a 750 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

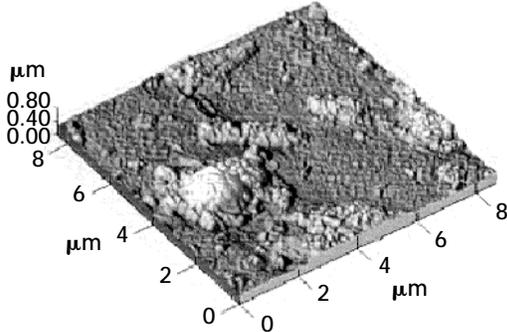


Figure 4.—Atomic Force Microscopy (AFM) micrograph showing the topology of the surface for the STO layer deposited at 750 °C.

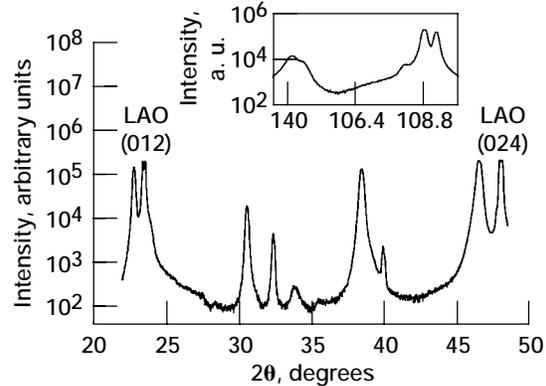


Figure 5.— 2θ - ω scan for the STO layer deposited at 750 °C.

STO film was single phase and epitaxial; a 2θ - ω scan for this film is shown in Figure 5. A $T_c=87.30$ K was measured for the YBCO film after fabrication of the multilayer structure. Since typical T_c 's for YBCO films deposited at 800°C are found to be between 88-90 K, any degradation of the YBCO due to the STO deposition was rather small.

Decreasing the STO growth temperature to 650°C, resulted in much lower ϵ_r values, lower tuning, and increased $\tan\delta$. Figure 6 shows that ϵ_r increased with decreasing temperatures, indicating that crystalline STO was formed, and was the material which dominated the electrical behavior. However, the low ϵ_r values indicate that the STO was poorly crystallized or that impurity phases with low ϵ_r were also present resulting in an overall lowering of the ϵ_r values. In the absence of an applied electric field, the $\tan\delta$ versus temperature data exhibit values of $\tan\delta$ slightly larger than those observed for the STO film deposited at 750°C. Furthermore, $\tan\delta$ rises sharply when an external dc bias is applied (see Figure 7). Figure 8 shows an AFM image of the film deposited at 650°C. The surface characteristics of this film are similar to those exhibited by the film deposited at 750°C, with a rms roughness of 40 nm and outgrowths near 140 nm. A difference between the 750 and 650°C films is that the density of outgrowths is higher for the 650°C sample than for the

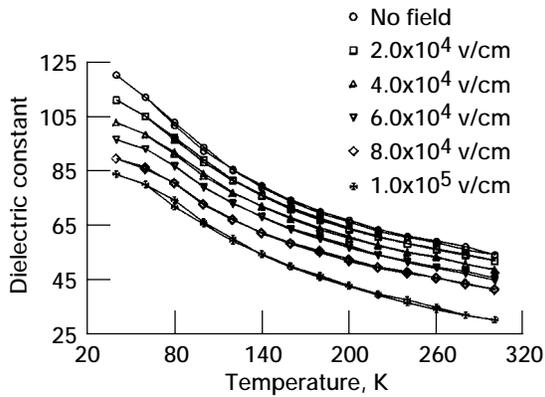


Figure 6.—Relative dielectric constant as a function of temperature and electric field for a 650 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

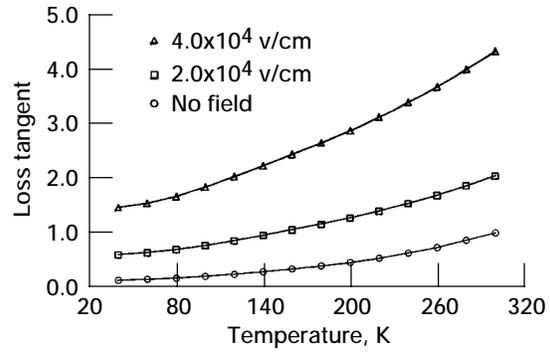


Figure 7.—Loss tangent as a function of temperature and electric field for a 650 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

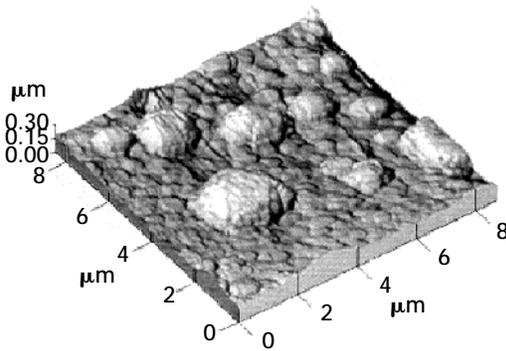


Figure 8.—Atomic Force Microscopy (AFM) micrograph showing the topology of the surface for the STO layer deposited at 650 °C.

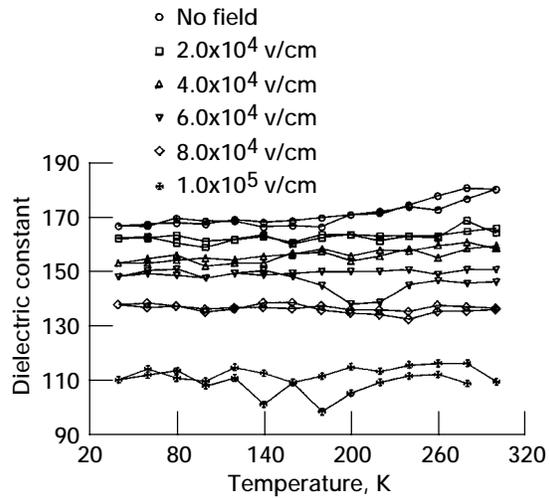


Figure 9.—Relative dielectric constant as a function of temperature and electric field for a 550 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

sample deposited at 750°C. A $T_c=83.79$ K was measured for the YBCO film after deposition of the STO layer at 650°C.

Lowering the STO growth temperature to 550°C resulted in a film with a higher ϵ_r than that observed in the film deposited at 650°C, but there was no well-defined temperature dependence to the ϵ_r (see Figure 9). In the absence of an applied field the values of $\tan\delta$ were somewhat lower than those of the film deposited at 650°C and slightly higher than those of the 750°C sample. However, similar to the 650°C sample, the $\tan\delta$ for the 550°C film increased sharply under the influence of an electric field (see Figure 10). A $T_c=85.82$ K was measured for the YBCO film after deposition of the STO layer at 550°C.

STO films deposited at temperatures from 250-450°C showed virtually no tuning, and $\tan\delta$ was high when a dc bias was applied. Figure 11 shows ϵ_r versus temperature data for an STO film deposited at 350°C. The ϵ_r values are low and drop with lower temperatures. As shown in Figure 12, increasing the bias across the films caused the $\tan\delta$ to increase sharply.

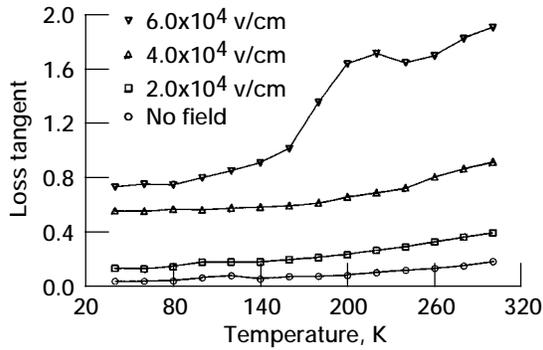


Figure 10.—Loss tangent as a function of temperature and electric field for a 550 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

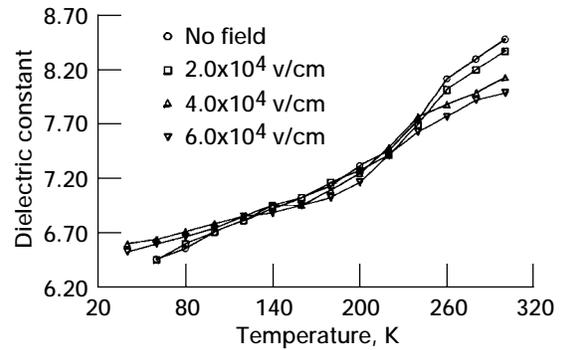


Figure 11.—Relative dielectric constant as a function of temperature and electric field for a 350 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

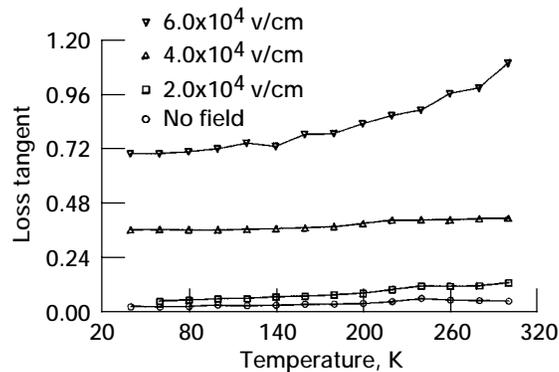


Figure 12.—Loss tangent as a function of temperature and electric field for a 350 °C deposited STO layer in a STO/YBCO/LAO multilayer structure. The data were taken at 1.0 MHz.

DISCUSSION

The only STO films in this study which displayed the electrical characteristics expected for high quality STO films were the films deposited at 750°C. The increase in ϵ_r with decreasing temperature and drop in ϵ_r when a dc bias is applied are similar to the trends observed in single crystals of STO¹², with the exception that bulk STO is an incipient ferroelectric and thus a peak in ϵ_r as a function of temperature is not observed¹³ contrary to the behavior observed in thin films. The high $\tan\delta$ observed in these films at temperatures above 90 K and the observed decrease as the temperature was lowered below the superconducting transition temperature, indicate that the $\tan\delta$ at high temperatures is primarily due to resistive losses in the YBCO film, and as the temperature is lowered these losses become negligible and the losses are dominated by the STO film. At 60 K, the increase in $\tan\delta$ for biases up to 4 volts indicates that the film is supporting electric fields up to 8×10^4 V/cm, and the small decrease in $\tan\delta$ with higher voltages indicates that the sample is nearing the onset of voltage breakdown. By contrast, interdigital capacitors comprised of highly

oriented STO films and a metallization arrangement in which both terminals are located on the same surface of the film reach voltage breakdown at fields over 10^5 V/cm and exhibit losses of less than 0.01.¹⁴ Thus we attribute the low breakdown voltages observed in the planar capacitors to electric field concentration at outgrowths and asperities in the STO films, which minimizes the voltage required for breakdown. Efforts are underway in our laboratory to produce smoother films so that the losses in the parallel plate configuration correlate better with those observed in the interdigital capacitors.

The STO film deposited at 650°C exhibited lower ϵ_r values, reduced tunability, and significantly higher losses with an applied field than those observed for the films deposited at 750°C . The fact that the ϵ_r increases with decreasing temperatures indicates the presence of the STO phase in the film, but the overall reduction of ϵ_r is a result of additional phases with low ϵ_r values. Unfortunately, quantification of the extent of STO formation using XRD is difficult because of overlapping between the STO and YBCO peaks in the XRD's 2θ - ω scan. At 60 K, $\tan\delta$ increases sharply with dc bias indicating that the breakdown voltage is lowered considerably by reducing the deposition temperature.

The ϵ_r values for films deposited at $T \leq 550^\circ\text{C}$ showed little temperature dependence, indicating negligible formation of the STO phase. At 60 K, the ϵ_r values for these films varied widely and therefore it is difficult to explain such behavior in terms of insulating dielectrics. We believe that since the breakdown voltages were readily exceeded in these films, the ϵ_r behavior was primarily controlled by free carrier transport and charge trapping at the electrode/STO interface or at defect sites such as grain boundaries in the STO films.

AFM data show similar surface roughness and grain size for films deposited at 550 - 750°C . Therefore, the difference in electrical behavior can not be attributed to the charge transport along microscopic defects such as debris from the target. Films deposited at 750°C have less outgrowths and exhibit highly faceted grains, indicating textured growth of the STO films. For films deposited at 650°C and 550°C the grains are more rounded, indicating less texture, and also exhibit a higher density of outgrowths than their 750°C counterpart. AFM images of films deposited at $T \leq 450^\circ\text{C}$ were blurred because particles on the surface of the films dislodged when the AFM needle was scanned across the film. This indicates that the films were granular and did not adhere well to the substrate.

CONCLUSIONS

Deposition of SrTiO_3 thin films with high ϵ_r values, large tunability, and high breakdown voltages requires the film to be deposited at elevated temperatures. At 60 K, the ϵ_r of film deposited at 750°C decreased by 35 percent when an electric field of 1×10^5 V/cm was applied. The $\tan\delta$ of the film deposited at 750°C increased slightly with dc bias, and increased sharply with dc bias for all films deposited at $T \leq 650^\circ\text{C}$. STO films deposited at 650°C exhibited reduced ϵ_r values and tunabilities, but the increase in ϵ_r with decreasing temperature suggest that there was some formation of STO. We conclude that for tuning applications, the growth temperature of STO films deposited on YBCO must be near 750°C so as to insure high voltage breakdown strengths in the films. Finally, a frequency dependence study of these structures may be helpful in distinguishing between extrinsic effects, such as YBCO/STO and Au/STO electrode effects, and intrinsic ones related to conduction through the STO film.

REFERENCES

1. V. K. Varadan, D. K. Ghodgaonkar, V. V. Varadan, J. F. Kelly, and P. Glikerdas, Microwave Jour., **35**, 116 (1992).
2. H. N. Al-Shareef, B. A. Tuttle, W. L. Warren, D. Dimos, M. V. Raymond, and M. A. Rodriguez, Appl. Phys. Lett., **68**, 272 (1996).
3. V. V. Lemanov, S. T. Pavlov and I. S. Pivovarov, Ferroelectrics, **144**, 101 (1993).
4. F. A. Miranda, C. H. Mueller, C. D. Cabbage, and K. B. Bhasin, IEEE Trans. Appl. Supercond., **5**, 3191–3194 (1995).
5. F. A. Miranda, C. H. Mueller, G. A. Koepf, and R. M. Yandrosky, Supercond. Sci. Technol., **8**, 755–763 (1995).
6. F. A. Miranda, K. B. Bhasin, K-S Kong, T. Itoh, and M. A. Stan, IEEE Microwave Guided Wave Lett., **2**, 287 (1992).
7. F. A. Miranda, S. S. Toncich and K. B. Bhasin, Microwave Opt. Tech. Lett., **6**, 752 (1993).
8. J. A. Beall, R. H. Ono, D. Galt, and J. C. Price, IEEE MTT-S Digest, 1421 (1993).
9. H-D Wu, Z. Zhang, F. Barnes, C. M. Jackson, A. Kain, and J. D. Cuchiario, IEEE Trans. Appl. Supercond., **4**, 156 (1994).
10. A. M. Hermann, A. Naziripour, A. Outzourhit, and C. H. Mueller, in Low Temperature Electronics and High Temperature Superconductivity, edited by C. L. Claeys, S. I. Raider, R. K. Kirschman, and W. D. Brown (The Electrochemical Society, New Jersey, 1995) pp. 35–49.
11. E. J. Tarsa, E. A. Hachfeld, F. T. Quinlan, J. S. Speck, and M. Eddy, Appl. Phys. Lett., **68**, 490 (1996).
12. R. C. Neville, B. Hoeneisen and C. A. Mead, J. Appl. Phys., **43**, 2124 (1972).
13. F. Gervais, in Handbook of Optical Constants of Solids II, edited by E. Palik (Academic Press, London, 1991), pp. 1035–1047.
14. C. H. Mueller, R. E. Treece, T. V. Rivkin, and J.B. Thompson, unpublished data.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE April 1996	3. REPORT TYPE AND DATES COVERED Technical Memorandum	
4. TITLE AND SUBTITLE Effect of SrTiO ₃ Deposition Temperature on the Dielectric Properties of SrTiO ₃ /YBa ₂ Cu ₃ O _{7-δ} /LaAlO ₃ Structures			5. FUNDING NUMBERS WU-233-5A-5A	
6. AUTHOR(S) F.A. Miranda, C.H. Mueller, R.E. Treece, T.V. Rivkin, J.B. Thompson, H.R. Moutinho, M. Dalberth, and C.T. Rogers				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration Lewis Research Center Cleveland, Ohio 44135-3191			8. PERFORMING ORGANIZATION REPORT NUMBER E-10135	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration Washington, D.C. 20546-0001			10. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA TM-107176	
11. SUPPLEMENTARY NOTES Prepared for the Eighth Annual International Symposium on Integrated Ferroelectrics, sponsored by the University of Colorado, Tempe, Arizona, March 17-20, 1996. F.A. Miranda, NASA Lewis Research Center; C.H. Mueller, R.E. Treece, and J.B. Thompson, Superconducting Core Technologies, 720 Corporate Circle, Golden, Colorado 80401; H.R. Moutinho, National Renewable Energy Laboratory, 1617 Cole Boulevard, Golden, Colorado 80401; M. Dalberth and C.T. Rogers, University of Colorado, Boulder, Colorado 80309. Responsible person, F.A. Miranda organization code 5620, (216) 433-6589.				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Unclassified - Unlimited Subject Category 76 This publication is available from the NASA Center for AeroSpace Information, (301) 621-0390.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) We report on the effect of the deposition temperature of SrTiO ₃ on the dielectric properties of SrTiO ₃ /YBa ₂ Cu ₃ O _{7-δ} /LaAlO ₃ thin film multilayer structures. In these structures, the YBa ₂ Cu ₃ O _{7-δ} (YBCO) films were deposited at 800°C by laser ablation, followed by the in-situ deposition of the SrTiO ₃ (STO) layer at one of the following temperatures: 750°C, 650°C, 550°C, 450°C, 350°C, and 250°C. Gold (Au) films were deposited and patterned on top of the STO layer to form planar Au/STO/YBCO capacitor structures. The electrical response was studied by measuring the dielectric constant (ε _r) and loss tangent (tanδ) of the ferroelectric film from 300-40 K, at 1.0 MHz, and at electric fields up to 100 kV/cm. Our results show 750°C to be a deposition temperature which allows for large variations of ε _r with limited enhancement of tanδ, while lower deposition temperatures cause a reduction of the induced change in ε _r and an increase in tan δ with applied field.				
14. SUBJECT TERMS SrTiO ₃ /YBa ₂ Cu ₃ O _{7-δ} /LaAlO ₃ multilayer structures; Thin films; Dielectric constant loss tangent; Deposition temperature			15. NUMBER OF PAGES 10	
			16. PRICE CODE A02	
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT	