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Off Axis Growth of Strontium Titanate Films with High Dielectric Constant Tuning and Low Loss

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ABSTRACT

We have measured the nonlinear dielectric properties of strontium titanate (STO) thin films grown on neodymium gallate (NGO) and lanthanum aluminate (LAO) substrates. The films prepared by off-axis pulsed laser deposition were characterized by their dielectric constant and loss tangent at 1 MHz and 2 GHz, and from room temperature down to 4 K. The resulting films show significant variations of dielectric properties with position of the substrates with respect to the plume axis. STO films on LAO substrates show low loss and high dielectric constant in regions near the plume axis. On the other hand, STO on NGO shows this effect only on the films grown far from the plume axis. We also obtained a figure of merit from the relative variation of the dielectric constant divided by the loss tangent in the presence of a DC electric field up to ± 4 V/ μ m. Careful mapping of the plume crosssection allowed us to improve the quality and reproducibility of the dielectric films, obtaining a best figure of merit at 2 GHz and 4 K close to 100 for NGO substrate but only well off axis.

INTRODUCTION

The dielectric constant of strontium titanate (STO) is a strong function of the applied electric field [1]. Thus, it is a promising material for fabricating RF and microwave devices such as phase shifters, tunable filters and dielectric varactors [2]. Also, the lattice mismatch between STO and the high temperature superconductors such as $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is on the order of 2% or better. This property as well as its good chemical compatibility make STO a good candidate for superconducting microwave electronics. The dielectric response of thin films shows a significantly different behavior than is seen for bulk STO. Unstressed bulk strontium titanate is an incipient ferroelectric material and it remains paraelectric down to the lowest temperature due to quantum fluctuations [3]. The dielectric constant of bulk STO increases nonlinearly from 300 at room temperature to 30000 at 4 K. On the other hand, thin films of STO, while showing a dielectric constant close to 300 at room temperature, typically reach a maximum between 1000 and 10000 in the 30 K to 100 K range depending on the film. Thin STO films also show a loss tangent which is an order of magnitude higher than the 10^{-3} - 10^{-4} of bulk STO. The microwave applications are hindered by the high dielectric loss in the material. Defects arising from lattice mismatch between the substrate and the film are believed to cause some of the loss, but other film defects could be involved [4]. Recently, approaches to produce films with reduced loss involved epitaxial lift off to relieve the strain in the films [5,6,8,9]. This method requires the growth of a sacrificial intermediate layer that needs to be etched away in order to release the film, hence involving a number of steps.

In this paper, we show that the pulsed laser deposition (PLD) growth conditions in different locations with respect to the plume axis can be chosen to produce good quality films in a single

growth step, thus avoiding a lift off technique. We report dielectric properties measurements performed on the films prepared with the off axis growth.

EXPERIMENTAL

Pulsed laser deposition (PLD) was used to deposit STO films on lanthanum aluminate and neodymium gallate substrates. This method has been used by a number of groups to prepare high quality STO films. The choice of the substrates was based on the lattice parameters in order to minimize the mismatch with STO (lattice parameters 0.3905 nm for STO (100), and roughly 0.379 nm and 0.385 nm for LAO (100) and NGO (110) respectively). A KrF excimer laser (1.8-2.3 J/cm², 246nm, 4Hz) was directed onto a rotating STO target (single crystal, purity 99.9%). The substrates were cut from commercial single crystal wafers, ultrasound cleaned in acetone and methanol and washed with distilled water. Cleaned substrates were mounted on a heating stage and placed in a vacuum chamber with an oxygen background pressure of 600 mtorr. The temperature of the stage was ramped from room temperature to 820 C, and maintained thereafter, until the desired film thickness was obtained. We obtained these optimized growth conditions from previous studies in our laboratory [7]. After the growth, the films were cooled down under 600 torr of oxygen pressure to avoid oxygen vacancies in the films. Ellipsometry was used to measure the film thickness, which was varied from 300-900 nm.

In order to perform dielectric constant measurements, Au coplanar capacitor electrodes were fabricated with the 25 micron gap separation, 1.5 mm total gap width and with overall 1x2 mm size. The capacitors were patterned on the surface of the films using a standard photolithographic technique, and were diced individually to 1x2 mm size. Capacitance measurement were taken with a network analyzer (HP 8510) for microwave frequencies, and with a LCR meter (HP 4275A) in the low frequency range, using a previously described ring resonator technique [6]. The capacitors were mounted in a temperature controlled ring resonator.

RESULTS AND DISCUSSION

In figure 1, (left panel) we compare the dielectric function at 1 MHz for STO films grown on LAO for different locations on the heating stage. Our heater stage is located 8.7 cm from the ablation target. The figure is labeled with the radial distance from where the plume center hits the heater stage. The thickness obtained from ellipsometry measurements is also shown in the figure 1. Notice that the films grown near the center of the plume show the highest values up to 8000 for the dielectric function. However, the exact position of the maximum is not at the plume axis. We also obtained data for the loss tangent shown on the right panel on figure 1. The loss tangent for all these films is lower than 0.015, which is significantly lower than the losses reported to date for STO films.

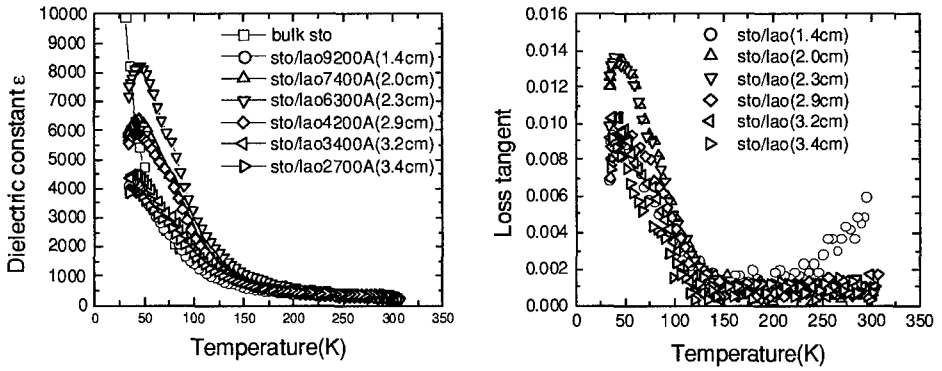


Figure 1. Dielectric constant and loss tangent at 1 MHz as a function of temperature of films STO grown on LAO at different locations with respect to the plume axis.

Similarly, figure 2 shows the dielectric function (left panel) and the loss tangent (right panel) at 1 MHz for STO films on NGO. The loss is also very small and, comparable to those of the STO films on LAO substrates. Films grown on NGO at the plume center have very poor dielectric response ($\epsilon < 1000$) and are not shown. Also from these observations, some of the films show an increase in their loss tangent at higher temperatures probably due to conductive loss.

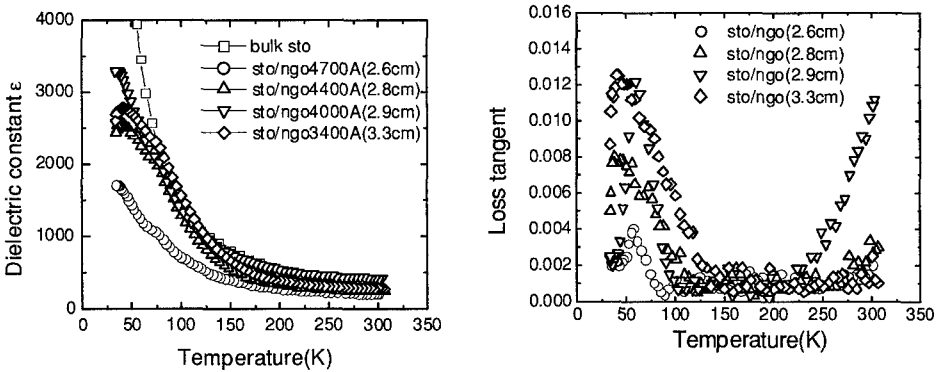


Figure 2. Dielectric constant and loss tangent at 1 MHz as a function of temperature of films STO grown on NGO at different locations with respect to the plume axis.

Figure 3 (left panel) shows the effect of substrate locations with respect to the plume axis on the maximum dielectric constant and loss tangent. The maximum of dielectric constant and loss tangent for films grown on different substrates mounted different location is plotted on the same graph. Notice that the films on LAO that sampled regions near the plume axis appear to have the higher maxima for the dielectric constant and the loss is low comparable to other films grown on edge of plume axis. On the other hand, the films on NGO mounted toward the edge of the plume have the higher maxima for the dielectric constant compared to the films on NGO mounted closed to the plume axis. The maximum for dielectric constant of films on LAO is higher than that of films on NGO.

To take the competing effects of tunability and loss into account, the figure of merit (K_E) is defined as the derivative of dielectric constant with respect to electric field divided by the product of dielectric constant and loss tangent. Figure 3 (right panel) shows the obtained figure of merit for different STO films. We observed that the figure of merit STO films grown on LAO and NGO have the maxima around 1000 at 1 MHz and 35 K, but only when the films are grown far from the plume axis. Thus, although the highest dielectric constant on LAO occurs near the plume center, the best tuning figure actually occurs about 3 cm off axis and at the same location as for optimal NGO based films. Based on the resulting figure of merit at 1 MHz and 35 K, the best films can be obtained by off axis growth.

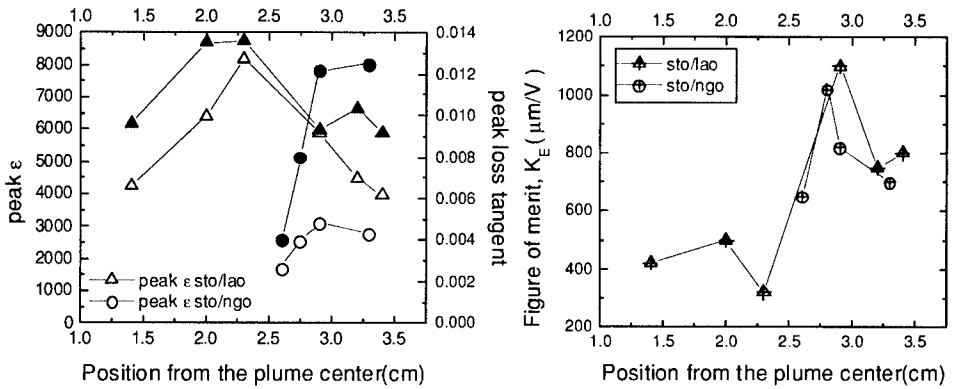


Figure 3. Effect of LAO and NGO substrate locations with respect to the plume axis 0 cm on the maximum of dielectric constant and loss tangent at 1 MHz ; open triangle- dielectric constant of STO on LAO, solid triangle- loss tangent of STO on LAO, open circle- dielectric constant of STO on NGO, solid circle- loss tangent of STO on NGO.

Finally, we have chosen one of the best films grown on NGO at 2.9 cm from the plume axis to study the microwave response. Figure 4 shows the dielectric constant and loss tangent as a function of temperature down to 4 K of this film measured at 2 GHz. The obtained peak dielectric is about 3000, and the loss tangent is less than 0.03.

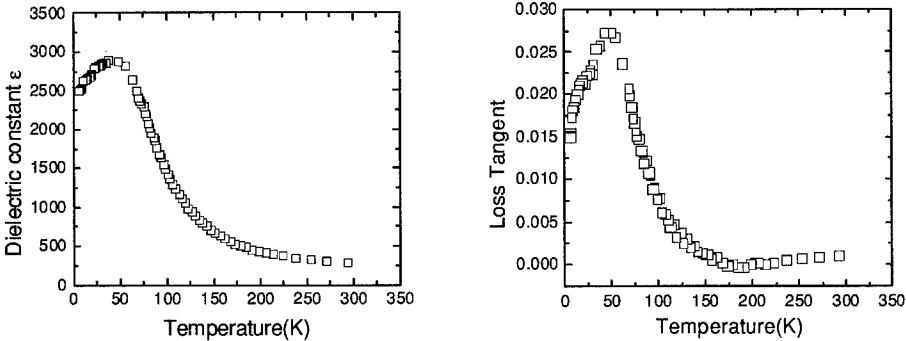


Figure 4. Dielectric constant and loss tangent at 2 GHz as a function of temperature of the best films STO grown on NGO at 2.9 cm from the plume axis.

With a DC electric field up to ± 4 V/ μ m, the film dielectric constant at 2 GHz and 4 K can be tuned by a factor of 4 and the loss is less than 0.015 as shown in figure 5 (right panel). Figure 5 (left panel) shows that the best obtained figure of merit at 2 GHz and 4 K is about 100. In the reference [6], the obtained figure of merit for films released from the growth substrate was 50. Thus, we have obtained twice the figure of merit while avoiding the epiaxial lift off method by using off axis PLD growth.

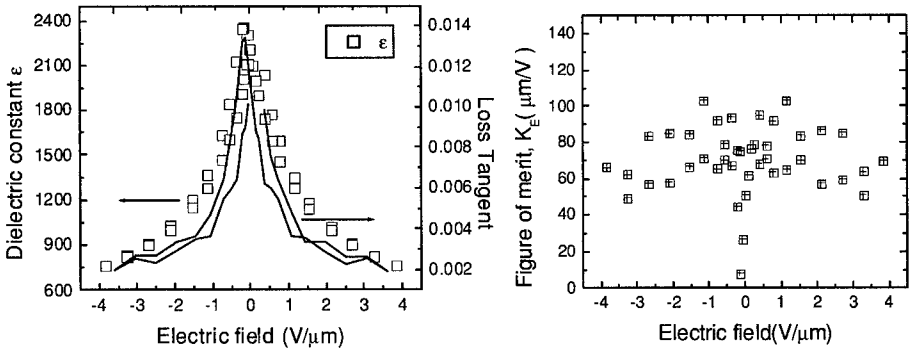


Figure 5. Tunability and figure of merit at 2GHz and 4 K of one of the best films on NGO.

CONCLUSIONS

STO films grown by PLD on different substrates at different locations with respect to the plume axis show significant variations of dielectric response. We observed that the STO films grown on LAO close to the plume axis has the maximum dielectric constant, while the same effect occurred on the films grown on NGO only far from the plume axis. The best figure of merit at 1 MHz and 35 K close to 1000 can be obtained when the films were grown on both substrates mounted roughly 3 cm from the plume axis. Also, we obtained a best figure of merit at 2 GHz and 4 K close to 100 for NGO substrate, but only for growth well off axis.

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