

## Dielectric properties of SrTiO<sub>3</sub> films under electric fields

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To obtain a high insulating STO layer, the parallel capacitors YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> / SrTiO<sub>3</sub> / YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> have been fabricated by a Chemical Mechanical planarization method. The epitaxial growth of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> and SrTiO<sub>3</sub> films were verified by X-ray diffraction, reflection high-energy electron diffraction and cross-section transmission electron microscopy. The dielectric constant of the SrTiO<sub>3</sub> film at 2.2 K was higher than 20000 at 100 kHz. The temperature variation of the dielectric constant fit the Curie-Weiss law above 150 K well and the Barrett formula within the temperature range 30 < T < 100 K. The best-fit parameters were similar to those for SrTiO<sub>3</sub> single crystals. Electric field dependence for the SrTiO<sub>3</sub>-film dielectric constant in parallel capacitors has been investigated at cryogenic temperatures. The dielectric constant was almost electric field independent at 2.2 K.

Key words: SrTiO<sub>3</sub>, YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>, Chemical Mechanical Planarization, Dielectric constant, Curie-Weiss law, Barrett formula, Electric field dependence

### 1. INTRODUCTION

For most high temperature superconducting devices such as the field effect transistor (FET),<sup>[1-5]</sup> a highly epitaxial insulation layer between superconducting films is necessary. To obtain a large magnitude of the electric field effect in FET's, both a high breakdown electric field  $E_d$  and a high dielectric constant  $\epsilon_r$  are required for the gate insulator.<sup>[3]</sup> To construct the epitaxial growth and obtain a superior insulation layer, SrTiO<sub>3</sub> (STO) is an appropriate material due to its small lattice mismatch (less than 2%) with YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> (YBCO).

STO is a quantum paraelectric material and its dielectric constant is almost independent of temperature within the cryogenic temperature range.<sup>[6-8]</sup> The dielectric constant of a STO single crystal can be higher than 20000 at around 2 K. However, the dielectric constant of STO films is usually rather low. Christen *et al.* reported that the dielectric constant of sputtered STO films was about 120 at 4.2 K and that the shift in the maximum value of the dielectric constant vs. electric field has been observed.<sup>[9]</sup> Li *et al.* showed that the dielectric constant of STO thin films grown on SrRuO<sub>3</sub> electrode layers is ~1200 at temperatures lower than 50 K.<sup>[10]</sup> A better dielectric constant (~12700 at 4.2 K) has been obtained in step-flow grown STO thin films under a bias field.<sup>[11]</sup> To our knowledge, this is the highest value ever obtained in

STO thin films. However, even this value is only half of that of the STO single crystal. The seriously degraded  $\epsilon_r$  limits the applications of STO thin films and the development of STO films with higher dielectric constants is desired.<sup>[3, 12-14]</sup>

For the integrated high temperature superconductor devices such as for FET's, top and base electrodes are high temperature superconductors. In the case of base YBCO electrodes, the problem of superconducting microshorts due to precipitates growing on base YBCO films is often encountered. To obtain a high insulation STO layer, preventing the formation of the superconducting microshorts is important.<sup>[15, 16]</sup> We have shown that a Chemical Mechanical Planarization (CMP) method is effective in eliminating the superconducting microshorts and improving the insulation properties of STO films for parallel capacitors YBCO/STO/YBCO.<sup>[17]</sup> The resistivity of the CMP STO film is  $2 \times 10^6$  and  $4 \times 10^7$   $\Omega\text{cm}$  at 300 K and 77 K, respectively. In this study we reported our findings for STO films fabricated by the CMP method. It is found that the dielectric constant of the CMP STO film is not only as high as that for a STO single crystal, but also hardly depends on an electric field. Both properties may aid development of field effect transistors.

## 2. EXPERIMENT AND RESULTS

### 2.1 Fabrication and crystallinity

The parallel capacitors of YBCO/STO/YBCO are fabricated by using the CMP method. After the deposition of the capped STO/base YBCO bilayer and the patterning process, a thick STO film ( $\sim 1\mu\text{m}$ ) was deposited on the patterned bilayer and subjected to polishing by the CMP method. The sample was then annealed at high temperature to rearrange atoms near the surface. An insulating STO film and top YBCO electrode were subsequently deposited. The fabrication process is described elsewhere in detail.<sup>[17]</sup> The epitaxial growth of YBCO (001) and STO (100) has been confirmed by x-ray diffraction, reflection high-energy electron diffraction (RHEED) and cross-sectional transmission electron microscopy (TEM).<sup>[17]</sup> X-ray diffraction patterns show peaks for only the YBCO (001) and STO (100) orientations. RHEED patterns of the base YBCO, STO and top YBCO show clear spots indicating the epitaxial growth of the films. Figure 1 shows the cross-sectional TEM image of the interface region between the top YBCO and the insulating STO layer. Clear interface and lattice images of the top YBCO and insulating STO layers are observed. Epitaxial growth of the top YBCO is confirmed. The interface is not observed at the interface-region between the CMP STO and insulating STO. This result indicates that epitaxial growth was achieved between the CMP STO and the insulating STO layers despite carrying out the CMP process.

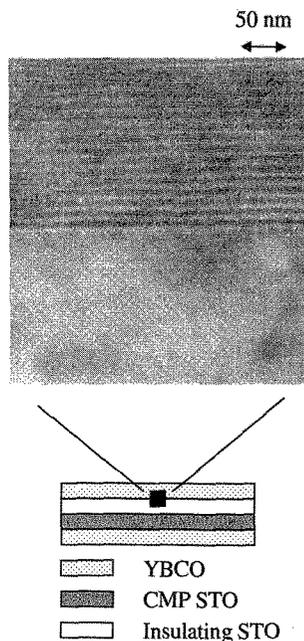


Figure 1 The cross-sectional TEM image of interface region between top YBCO and insulating STO layer.

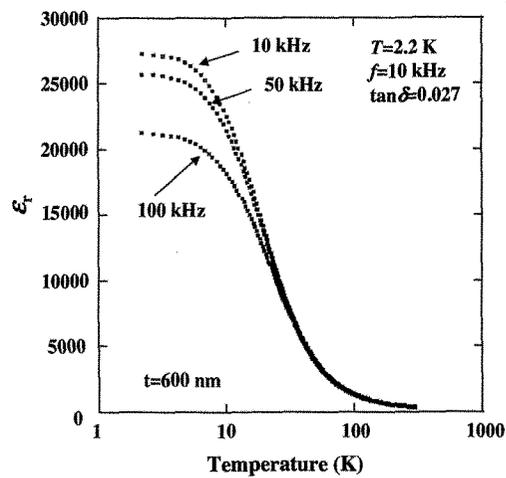


Figure 2 Temperature dependence of dielectric constant measured in parallel capacitors for a CMP STO film with a thickness of 600 nm.

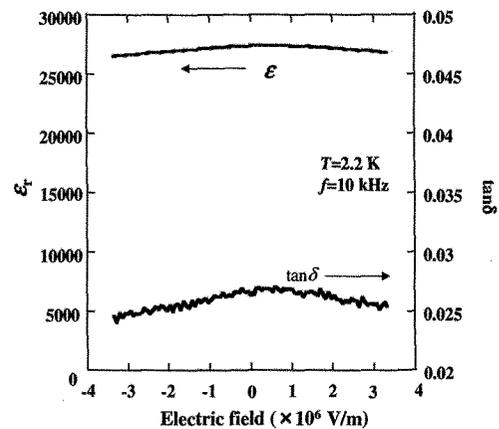


Figure 3 The dielectric constant under electric fields at 2.2 K and 10 kHz. The dielectric constant under electric fields between 0 and  $\pm 3.5 \times 10^6$  V/m are 27200 and 26500, respectively.

### 2.2 Dielectric behavior

Capacitance was measured by using an HP4284A precision LCR meter with an ac test signal amplitude of 5 mV. The dielectric constant  $\epsilon_r$  was deduced from the measured capacitance by using the relation  $C = \epsilon_0 \epsilon_r A / t$ , where  $A$  is the area of an electrode and  $t$  the thickness of the STO layer between both electrodes. Figure 2 shows the temperature dependence of the dielectric constant for a STO film with  $t = 600$  nm at 10, 50, and 100 kHz. Well-defined quantum paraelectric behavior is observed for the CMP STO film. In addition,  $\epsilon_r$  is  $\sim 330$  at room

temperature and  $\epsilon_r$  is  $\sim 21000$  (100 kHz) at 2.2 K, respectively. These values are very close to that of the STO single crystals. Above 150 K, the inverse dielectric constant fits the classical Curie-Weiss law:  $1/\epsilon_r \cdot \epsilon_0 = (T - \theta_c)/C$ , with the best-fit parameters  $C = 0.738 \times 10^{-6}$  FK/m and  $\theta_c = 33$  K in good agreement with that reported for STO single crystals.<sup>[17]</sup> In the temperature range  $30 < T < 100$  K, the data in Fig. 2 fits the Barrett formula,<sup>[18]</sup>  $\epsilon_r(T) = A/[T_1/2 \coth(T_1/2T) - T_0]$ , where  $T_0$  indicates the Curie-Weiss temperature, and  $T_1$  indicates the dividing point between the low temperature region where quantum effects are important and the high temperature region where a classical approximation is valid. The best-fit parameters are  $A = 0.85 \times 10^5$ ,  $T_1 = 87$  K and  $T_0 = 37$  K. These parameters are similar to those found in pure STO single crystals,  $T_1 = 84$  K and  $T_0 = 38$  K.<sup>[8, 19]</sup> It is well known that STO is very sensitive to impurities. Minor impurities or imperfections have a strong effect on its dielectric behavior and the value of  $\epsilon_r$ . The observed single crystal-like behaviors indicate that the CMP STO film is of high quality and free from impurities and imperfections. Usually, the degradation of  $\epsilon_r$  for STO thin films is attributed to the existence of dead layers at the electrode/dielectric interfaces.<sup>[10]</sup> The high value of  $\epsilon_r$  for the CMP STO film seems to suggest that the lattice for the STO film and the YBCO electrodes match well and have almost no influence on the dielectric properties. At temperatures lower than  $\sim 50$  K, the dielectric constant of the CMP STO film exhibits obvious frequency dependence. Furthermore, dielectric loss,  $\tan \delta$ , shows 0.027 at 2 K and 10 kHz.

Electric field ( $E$ ) dependence of the dielectric constant was measured at a frequency of  $f = 10$  kHz by using an HP4284A LCR meter. After the zero-field cooled to the measurement temperature, data was collected under isothermal conditions where the electric field was swept in the following sequence:  $0 \rightarrow E_{b,max} \rightarrow 0 \rightarrow -E_{b,max} \rightarrow 0$ , where  $E_{b,max}$  denotes the maximum bias field applied. For data evaluation, only the data taken in the middle of the sweep,  $E_{b,max} \rightarrow 0 \rightarrow -E_{b,max}$ , was used. Figure 3 shows the variation of the dielectric constant and  $\tan \delta$  versus electric field at 2.2 K.  $\epsilon_r$  is 27200 under  $E = 0$  and 26500 under  $E = \pm 3.5 \times 10^6$  V/m, respectively. The change of  $\epsilon_r$  was less than 3%. The shift of the maximum  $\epsilon_r \sim E$  curve was hardly observed. Also,  $\tan \delta$  is 0.027 under  $E = 0$  and 0.025 under  $E = \pm 3.5 \times 10^6$  V/m, respectively.

### 3. DISCUSSION

For STO parallel capacitors under  $E \sim 10^6$  V/m, the change in dielectric constant strongly depends on sample preparation.<sup>[9, 10]</sup> Lippmaa *et al* have found that for STO thin films grown on SrRuO<sub>3</sub> electrode layers,  $\epsilon_r$  could be tuned by 80% under electric fields  $\pm 3 \times 10^6$  V/m.<sup>[10]</sup>

However, Christen *et al* have shown that  $\epsilon_r$  of sputtered STO films is hardly affected by electric fields up to  $\pm 10^7$  V/m.<sup>[9]</sup> It has been suggested that the electric field dependence of  $\epsilon_r$  results from the nonlinear relation between the applied electric field and the polarization, and the shift of the maximum  $\epsilon_r \sim E$  curve is attributed to strong charge trapping at the film-substrate interface. For parallel capacitors with different bottom and top electrodes such as Pt/STO/Nb-doped STO, the shift of the  $\epsilon_r \sim E$  curve maximum was explained by the asymmetric work functions of the Nb-doped STO and Pt electrodes. For our CMP STO film, the field dependence of the dielectric constant is fairly weak. This result may suggest that the relation between the applied field and the polarization is linear for the CMP STO film. The weak shift of the maximum in dielectric constant versus electric field is observed. This might indicate that the influence of strong charge trapping at the film-substrate interface is very small for our CMP STO film. Evidently, the breakdown electric field  $E_d$  is higher than  $3.5 \times 10^6$  V/m for the CMP STO film.

### 4. CONCLUSION

Dielectric properties of the CMP STO (100) films on YBCO (001) have been investigated from room temperature to 2.2 K. Compared to the STO film fabricated by other methods, the CMP STO film is characterized by: a high dielectric constant comparable to that of a STO single crystal, a weak electric field dependence of the dielectric constant, and a small shift in maximum value of the dielectric constant vs. electric field. The large dielectric constants allow the transfer of a large number of charges with a moderate FET gate voltage and the weak electric field dependence may widen its applicability. Thus, it is possible that a high performance FET could be constructed with our CMP STO film.

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