Improved SrTiO$_3$ thin films using oxygen relaxation technique

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The oxygen relaxation technique for thin-film growth is explored. In this technique, films are deposited as nanoscale multilayered structures. After the deposition of each layer, the deposition process is stopped, the sample is cooled down slowly in situ in an oxygen-rich atmosphere to a temperature of 250 °C, then it is heated up to the set deposition temperature for growth of the next layer. The results show that by using the oxygen relaxation technique, it is possible to improve the electrical properties and the microstructure of SrTiO$_3$ films keeping the surface smooth and free from pinholes and pores. The oxygen relaxation technique is more efficient for smaller film thickness repetition. © 2005 American Institute of Physics. [DOI: 10.1063/1.2136432]

Strontium titanate (SrTiO$_3$, STO) thin films have been extensively studied for application in tunable microwave components. The most commonly investigated devices are varactors, phase-shifters, tunable filters, etc., where the STO films are used as a single layer or are included in multilayer structures.$^{1,2}$ The STO films’ undesirably high dielectric loss at microwave frequencies, however, limits their wide practical utilization. It is believed that one reason for the high microwave loss level is the oxygen deficiency in the STO films, and a postdeposition high-temperature annealing in oxygen atmosphere is required.$^3$ Very often, however, after such an annealing, pores and/or cracks appear on the film surface.$^4$ This is unacceptable because of the possibility of a short circuit between the layers (in multilayered structures).

In this letter, the oxygen relaxation technique for growing STO films with improved electrical properties is explored to investigate the dependence of the STO film quality on the oxygen relaxation treatment.

A set of STO films was on-axis deposited on 5 × 5 mm$^2$ (001) LaAlO$_3$ (LAO) substrates by laser ablation from a stoichiometric SrTiO$_3$ target. The film thickness of all samples was 400 nm, although they were made without (Sample 1), with one (Sample 2), with three (Sample 3), with five (Sample 4), and seven (Sample 5) intermediate oxygen relaxations, i.e., the films were deposited as nanoscale multilayered structure with one (1 × 400 nm), two (2 × 200 nm), four (4 × 100 nm), six (6 × 65 nm), or eight (8 × 50 nm) layers. After deposition of every layer, the deposition process was stopped, the sample cooled down slowly (20 °C/min) in an oxygen-rich (0.9 atm) atmosphere to a temperature of 250 °C, then heated up to the deposition temperature for growth of the next layer.

For examination of electrical properties, the deposited STO films were covered by (250 nm) YBa$_2$Cu$_3$O$_{7-x}$(YBCO)/(500 nm)Au bilayer. The YBCO layer was in situ laser ablated, while the Au one was ex situ electron-beam evaporated. The deposition conditions for the STO, YBCO, and Au films can be found elsewhere.$^5$ The test structures (planar capacitors) for electrical measurements were patterned by photolithography and Ar-ion milling.

The sample structure was investigated by x-ray diffraction (XRD) performed on a Philips X’Pert PW3098/20 diffractometer with Cu $K\alpha$ radiation. A Veeco scanning probe microscope (SPM) in tapping mode was used for investigation of the STO film surface.

Electrical measurements of the planar capacitors were made at a radio frequency (rf) of 2 MHz and at high frequency (MW) of 6 GHz, at both 300 K and 77 K. To examine the capacitors’ tunability $n=C(0 V)/C(V_{\text{max}})$, a dc voltage up to 40 V was applied. Measurements at 77 K were performed by immersing the capacitor in liquid nitrogen. The rf measurements were performed using a HP 4285A LCR meter attached to a microwave probe station. Before measurement, a calibration using an open and short circuit was performed. For MW measurements, the substrate was cut and the chip capacitors were placed into a gap in the middle of the half-wavelength microstrip resonator. The measurement and parameter evaluation procedures are described in Ref. 6.

The XRD patterns measured for all YBCO/STO/LAO structures showed only (001) reflections of YBCO and STO. A typical XRD pattern is presented in Fig. 1. After a broadening analysis of the obtained XRD patterns, the strain value for all samples were estimated as $|c_f-c_{\text{bulk}}|/c_{\text{bulk}}$, where $c_f$ is the lattice parameter along the film thickness, and $c_{\text{bulk}}$ is the lattice parameter for a bulk STO sample. The strain gradient was estimated as $\Delta c/c_f$, where the $\Delta c$ is the deviation of the STO lattice parameter the along the film thickness.

![FIG. 1. Typical XRD pattern of the YBCO/STO/LAO multilayered structure.](image)

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from the average STO thin-film lattice parameter. More about XRD measurements can be found elsewhere.\(^7,8\) A measurements summary is presented in Table I. One can see that increasing the number of oxygen relaxation reduces the strain gradient, while the strain value \((|c_f - c_{\text{bulk}}| / c_{\text{bulk}})\) is more or less constant. It is difficult to provide a strong explanation of the observed phenomenon but one can speculate that, after oxygen relaxation procedures, the STO film does not undergo additional recrystallization, which takes place during the conventional annealing, but becomes a homoepitaxial multilayered structure. The fact that the strain value remains the same after different numbers of oxygen relax-

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Number of oxygen Relax.</th>
<th>Crystal cell parameters</th>
<th>Cryo temperature (77 K)</th>
<th>Room temperature (300 K)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(c_f ) (\AA)</td>
<td>(</td>
<td>c_f - c_{\text{bulk}}</td>
<td>/ c_{\text{bulk}})</td>
</tr>
<tr>
<td>1</td>
<td>w/o</td>
<td>3.916</td>
<td>0.0028</td>
<td>0.0012</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>3.916</td>
<td>0.0028</td>
<td>0.0009</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>3.916</td>
<td>0.0027</td>
<td>0.0009</td>
</tr>
<tr>
<td>4</td>
<td>5</td>
<td>0.916</td>
<td>0.0026</td>
<td>0.0006</td>
</tr>
<tr>
<td>5</td>
<td>7</td>
<td>3.915</td>
<td>0.0026</td>
<td>0.0002</td>
</tr>
</tbody>
</table>

FIG. 2. SPM images of the STO film surface measured for: (a) Sample No. 1 (without oxygen relaxation); scale in \(z\) direction is 50 nm/div. and (b) Sample No. 5 (with seven oxygen relaxations); scale in \(z\) direction is 25 nm/div.

FIG. 3. Typical \(C-V\) and \(\tan \delta\)-voltage characteristics measured at (a) 77 K, 2 MHz, and (b) 77 K, 6 GHz.
ations supports the idea that there is no additional recrystal-

lization of the film.

The change in the strain gradient can be explained as
follows: Initially, we deal with a strained STO film in which
the lattice parameter changes along the thickness. The oxy-
gen relaxation makes the sample consisting of similar homo-
epitaxial layers, in which, the strain gradient (dispersion
of the lattice parameter) is smaller. Another interesting fact is
that oxygen relaxation at every 50 nm reduces the strain gra-
dient inside a 400 nm STO film 83%, while one at 65 nm
reduces the strain by 50%, showing that the oxygen relax-
tion technique is more efficient for smaller film thickness
repetition.

In Fig. 2 are shown typical surface images obtain by
SPM for Sample Nos. 1 and 5. As one can see, the surface is
very smooth and free from pinholes and pores, which usually
take place after a postdeposition high-temperature annealing.
This is important especially for multilayer-based device
fabrication.

Results of the rf/MW measurements are also summa-
rized in Table I. Presented there are the capacitance ($C$),
the tuning ($n$), and the $Q$ factor ($Q = 1/\tan \delta$) of the capacitors
measured at 2 MHz and 6 GHz and at 77 K and 300 K. The
improvement of the electrical properties of the films depos-
itmed using the proposed technique is clearly seen at low fre-
cuencies, where the $Q$ increases approximately five fold. At
high frequencies, the improvement is within the measure-
ment error, which does not exceed 5–10%. The increase in
capacitance observed at rf and MW may be attributed to the
samples layered structure. (The layered sample with planar
capacitor design can be considered as one that consists of
few capacitors connected in parallel).\(^9\)

Figures 3(a) and 3(b) show typical capacitance-voltage
(C-V) and $\tan \delta$-V dependences, measured at 77 K at
2 MHz and 6 GHz, respectively. The weak hysteresis loop
that takes place, especially at low frequencies, is attributed to
the redistribution of the space charge in the underelectrode
area or/and strain-induced ferroelectric domains in the STO
films.\(^10\) As expected, no tuning was detected at
room temperature.

In conclusion, the investigation shows that by using the
oxygen relaxation technique, it is possible to improve the
structural and the electrical properties of the STO films keep-
ing the surface smooth and free from pinholes and pores. The
capacitance of the structure increases proportionally to the
number of the oxygen relaxations. The same trend is ob-
erved at rf, where the capacitors’ $Q$ factor increases about
five times, while at microwave frequency the improvement is
within the measurement error. Oxygen relaxation repeated at
every 50 nm reduces the strain gradient of a 400 nm STO
film 83%. Increasing the repetition layer thickness reduces
the efficiency of the oxygen relaxation technique.

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