AFM study of SrTiO$_3$ and YBa$_2$Cu$_3$O$_{7-\delta}$ multilayer surface treated with chemical mechanical polishing process

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Abstract. We have studied the surface morphology of SrTiO$_3$ (STO) and YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thin films fabricated by pulsed laser deposition technique in a multilayer structure. Study was focused on the surface morphology of an STO thin film deposited on an YBCO layer that received chemical and mechanical polishing (CMP) treatment. The CMP process could remove the presence of the outgrowth on the STO thin film, improve its smoothness and facilitate a good epitaxial growth of the next deposited YBCO film. As a result, we could fabricate a high quality YBCO thin film on the YBCO/STO structure with $T_c = 91$ K and $J_c = 4 \times 10^6$ A/cm$^2$.

1. Introduction
Combination of superconductor and other materials such as insulator, dielectric, magnetic, etc. in the multilayer structure has potential application as devices. We could mention those devices such as SQUID [1], field effect transistor (FET) [2], spin injection device [3], capacitor [4]. The multilayer structure consisting of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) and SrTiO$_3$ (STO) layer is extensively studied for the fabrication of SQUID with flux transformer [4] and microwave device [5]. It is because the two materials have low lattice parameter mismatch, which allows a good epitaxial growth of both materials.
The pulsed laser deposition (PLD) technique [6] is widely used in the fabrication of oxide thin films since this technique is simple and versatile. With PLD technique, we could fabricate thin films with similar chemical and mechanical properties as those of the target. However, this method also has disadvantages such as difficulties in the fabrication of large area thin films and the medium quality of the surface of the fabricated thin films. Factors that affect the quality of thin film surfaces are the presence of outgrowth, precipitation or droplet and non-uniform growth of the grains on the film and these would raise the roughness of the film surface. It will prevent the epitaxial growth of the next deposited film. Here we study the application of the chemical mechanical polishing technique to improve the smoothness of the STO film surface grown on an YBCO layer in order to facilitate a good epitaxial growth of additional YBCO layer. So far, CMP method was used in the multilayer devices to overcome the step-edge problem and to increase the device density in the integrated circuit [7, 8].

2. Experimental
YBCO and STO thin films are prepared by pulsed laser deposition technique using an ArF excimer laser ($\lambda = 193$ nm). First, YBCO film was deposited on the STO (001) substrate heated at 745 °C with halogen lamp. The oxygen pressure was adjusted at 700 mTorr. The laser energy was set at 100 mJ
with a repetition rate of 2 Hz. The thickness of deposited YBCO films in this study was 300 nm. STO thin films grown on YBCO layers were prepared with the same conditions except that the laser energy and repetition rate were set at 200 mJ and 4 Hz, respectively. The thickness of the STO thin films was about 1 μm. We used an YBCO ceramic and a STO single crystal as targets.

Chemical mechanical polishing was performed with MA-200 polishing system of Musashino Denshi Co. Ltd. We used polishing material of soft felt M.M. 431 from Heraeus GmbH laid on the rotation table and diamond slurry with particle size of 0.5 μm dissolved in water and ethylene glycol (Hyprez S-4889) from Engis Co. Ltd. The rate of rotation of the table was set at 60 rpm.

Characterization of the film was carried out using the atomic force microscopy (AFM), RHEED and X-ray diffraction (XRD). We patterned the films into a bridge with a width of 20 μm and measured the superconducting properties (Tc and Jc) using a standard four-probe method.

3. Results and discussions

The AFM image of a typical YBCO film grown on an STO substrate is shown in Fig. 1(a). We can clearly see that the YBCO film has island growth with an average size of 40 x 40 nm². Hollows are also observed between the grains with a large variation in size. The nano-sized precipitates are also observed on the film. The root-mean-square (RMS) roughness for the film was 4.7 nm. The AFM image of a typical 1 μm-thick STO thin film grown on the YBCO is shown in Fig. 1(b) (denoted as sample 1). On this film, large outgrowths with an average diameter of 0.6 μm are observed. On the other area, some rectangular grains are observed with large distribution of its height. The RMS roughness value of this film was 53.2 nm. It is interesting to find a fabrication method that can reduce the presence of the outgrowth. We could prepare a thick STO film without outgrowth and its AFM image is shown in figure 1(c) (denoted as sample 2). This film was deposited subsequently after the deposition of YBCO film, meanwhile the previous one (sample 1) was prepared after the YBCO film was annealed and then cooled to room temperature.

After a CMP process and ultrasonic cleaning in acetone were made for the sample 1, an AFM image of the film was taken and is shown in Fig. 2(a). The large outgrowths on the film were disappeared but a number of small amorphous structures are seen. These amorphous structures are the remnant of the rectangular STO grains. To remove these structures and rearrange the atoms on the STO surface, we performed an annealing process at 600 °C with oxygen pressure of 700 mTorr for 2 hours. It is clear that the amorphous structures disappeared as it is shown in Fig 2(b). There is a large improvement in the smoothness of the film, i.e. the STO thin film prior to the annealing process show an RMS roughness of 4.6 nm and after annealing it becomes 2.7 nm. We note that some scratches also occur during the CMP process. The depth of the scratches is around 7.5 nm, which is shallower than the hollow observed on the YBCO film grown STO single crystal.
It is important to identify the origin of the outgrowth found on the STO film. The XRD spectra of the as grown STO film on YBCO layer from both samples (1 and 2) are shown in Fig. 3. We found that in the XRD spectra taken from sample 1, there are peaks arising from the STO (110) and (111) reflection planes. The result shows that the outgrowth on the STO film is the agglomeration of the STO (110) and (111) oriented grains. On the other hand, the XRD spectra from the sample 2 show only the reflection from the (00/l) plane of STO substrate/thin film and (00/l) plane of YBCO.

The AFM images of the YBCO thin film grown on the STO layer that received CMP and annealing treatments are shown in Fig. 4. These YBCO films have thickness around 200 nm. On the sample 1 (Fig. 4(a)), YBCO thin film mainly consists of grains similar to the grains on the YBCO film grown
on the STO single crystal, which indicate a good epitaxial growth of the c-axis oriented grains. However, there are large and deep hollows on the film and these areas are the former sites of the large STO outgrowths. Since on that site there are different orientations of STO grains, a good epitaxial c-axis oriented grain could be difficult to grow. On the other hand, we observed smooth and well connected grains of YBCO in sample 2 (Fig. 4(b)) and show a good epitaxial growth of c-axis oriented grains.

Fig. 5. (a) Resistivity vs. temperature for YBCO thin film grown on the top of STO thin film, which received CMP treatment. (b) The critical current density vs. temperature. The blue line is from the sample 1 and the red one is from sample 2.

The superconducting critical temperature ($T_c$) of the YBCO thin films was shown in Fig. 5(a). For the YBCO thin film grown on an STO single crystal, the zero resistivity ($T_{c,0}$) was achieved at 92 K with the $\Delta T_c$ less than 1 K. For the top YBCO films, which have outgrowth observed on the STO layer (sample 1), the $T_{c,0}$ is about 91.2 K and the $T_{c,0}$ for the top YBCO film in sample 2 is found to be 91 K. However, both films have very sharp transition ($\Delta T_c < 1$ K). The superconducting critical current density, $J_c$ data are shown in Fig. 5(b). The YBCO layer grown on an STO substrate has $J_c$ around $3 \times 10^6$ A/cm$^2$ at 77 K. Meanwhile, the top YBCO layer of the sample 2 has $J_c$ of $4 \times 10^6$ A/cm$^2$ at 77 K, which is better than the bottom layer YBCO. However, the $J_c$ value at 77 K for the top YBCO thin film of sample 1 shows a value of $2.9 \times 10^5$ A/cm$^2$, which is almost 10 times lower that the other YBCO thin films. This could be understood since in the sample 1, large number of hollow present and they limit the area where the super-current could flow on the film.

4. Summary
We have studied the surface morphology of an YBCO thin film grown on the STO substrate, an STO thin film grown on YBCO layer, which was treated by the CMP process, and a top YBCO film deposited on the STO film. We found that on the STO layer large outgrowths could present and they could be removed by the CMP process. The CMP could increase smoothness of the STO thin film and facilitate a good epitaxial growth of the next deposited YBCO thin film. The quality YBCO film on the STO layer is characterized by high $T_c$ values and high $J_c$ values at 77 K that exceeds those of YBCO thin films grown on STO single crystals.

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