Epitaxial Growth and Characteristics of the YBCO/STO/YBCO Tunneling Junctions

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We report the growth on MgO (100) of the insulating SrTiO₃ (STO) thin film and the YBa₂Cu₃O₇₋ₓ(YBCO)/STO heteroepitaxial multilayer structure using the in-situ pulsed laser ablation technique. By simply placing a shadow mask between the target material and the substrate, the particulate problem peculiar to the excimer laser deposition technique was effectively eliminated. Epitaxial YBCO thin films on STO/MgO and STO/YBCO/MgO with Tc as high as 91 K and 84 K were obtained by this "eclipse method". We deduce the energy gap of YBCO from the tunneling characteristics (dI/dV-V) of our YBCO/STO/YBCO junctions as Δ₂=19-21 meV and Δ₁=4-5 meV. These results are consistent with the existing reports.

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I. Introduction

For the growth of thin film of oxide superconductors, pulsed laser deposition (PLD) technique has been used widely [1,2]. The PLD technique is advantageous over other existing deposition methods due to such factors as the high growth rate and the full transfer of the composition from target to film. However, it suffers from a significant problem of particles and/or droplets which scatter on the surface of the grown film. The defective morphology brought about by those anomalies is unfavorable for the microelectronic applications of the high-T, superconductors. Several attempts had been made to solved the aforementioned problem in PLD [3,4]. For example, Holzapfel et al. proposed an off-axis PLD in which the substrate has less chance to receive flying particles [5]. Recently, Kinoshita et al. carried out a PLD experiment in which a shadow mask was introduced into the pulsed laser deposition system, placing between the target and the substrate [6]. The radiation spot in the target (the sun) is screened by the shadow mask (the moon) from view on the substrate (the earth), and therefore, this method was called the eclipse PLD [6], in which the deposition proceeds through diffusion only.

In the present work, we report the growth on MgO of the superconducting YBa₂Cu₃O₇₋ₓ(YBCO) and the insulating SrTiO₃(STO) thin films and the fabrication of the YBCO/STO heteroepitaxy, using the in-situ eclipse PLD technique. The results were compared with those of the conventional (without the shadow mask) PLD.
II. Experimental

Due to the small mismatch in the lattice parameters and the thermal expansion coefficient between YBCO and STO, we chose STO as our buffer layer and insulating layer [7]. YBCO targets were prepared by conventional ceramic process, using the stoichiometric amounts of oxide powders of Y$_2$O$_3$, BaCO$_3$ and CuO. A XeCl pulsed excimer laser (308 nm, 10 ns pulses) was used for film ablation. The repetition rate was 5 Hz and the energy density on the rotating target was 1.5 J/cm$^2$. The target-substrate distance and the substrate-shadow mask distance were 35 mm and 25 mm, respectively, and the shadow mask size was 1 x 5 mm$^2$. MgO (100) substrates with the size of 2 mm x 6 mm x 0.5 mm were placed behind the shadow mask. The substrate temperature was 600 °C for the buffer STO layer, 560 °C for the bottom YBCO layer and the insulating STO layer, and 500 °C for the top YBCO layer. The ambient oxygen pressure during the deposition was maintained at 0.1 mbar for the buffer layer, 0.2 mbar for the insulating layer, and 0.4 mbar for the YBCO layers. The thickness of the film was typically 300 nm for the YBCO layers, 45 nm for the insulating layer and 15 nm for the buffer layer.

The sandwich-type S/I/S junctions were fabricated in-situ as follows: First, a bottom YBCO layer was deposited on the right half of STO-coated MgO (100) substrate through a set of adjustable metal mask. Next, the metal mask was moved to one end of the fixture such that the insulating STO layer was epitaxially grown on top of the bottom YBCO layer. Finally, the metal mask was moved to the other end and a top YBCO layer was deposited on the left half of the substrate on top of the insulating STO layer. The effective tunneling area, which was the overlapping area of the two YBCO layers, was about 0.5 x 2 mm$^2$.

The surface morphology of the deposited films was observed by scanning electron microscopy (SEM) and atomic force microscopy (AFM). X-ray diffraction (XRD) and energy dispersive spectroscopy (EDS) measurements were conducted for characterizing the grown films. The R-T curves and tunneling characteristics of our sample were measured at varied

![FIG. 1. SEM images from the YBCO films deposited by (a) conventional PLD and (b) eclipse PLD.](image)
FIG. 2. Characteristics of the eclipse-PLD grown YBCO film: (a) XRD pattern, (b) resistance vs temperature.

III. Results and discussion

Figures 1(a) and 1(b) are the SEM images from the YBCO films deposited by conventional and eclipse PLD, respectively. As shown in Fig. 1(a), the film contains many scattered particulates with size of 0.1 \(-\) 0.5 \(\mu\text{m}\). These particulates are generally thought to be introduced directly from the target or to be condensed during expansion in the ambient plume. In contrast with this conventionally deposited film, smooth and droplet-free surface morphology is observed for the film grown by eclipse PLD (Fig. 1(b)). The growth rate is reduced to 3 \(\text{nm/min}\), which is 50% of that in conventional PLD in the same ambient oxygen. The average roughness for the films grown by eclipse PLD is around 20 nm, while
FIG. 3. Three-dimensional AFM images of the STO/YBCO bilayers. The horizontal dimension is 1.7 μm for each graph. The thickness of STO is 60 nm for (a), 30 nm for (b), 15 nm for (c), and 3 nm for (d).

that for the films grown by conventional PLD is around 40 nm.

Typical XRD pattern from the YBCO film grown by eclipse PLD after the deposition of STO is shown in Fig. 2(a), indicating that the YBCO film is (OOI)-oriented. The YBCO film grown by eclipse PLD exhibits a metallic conducting property, as shown in Fig. 2(b). A zero-resistance temperature ($T_{CO}$) as high as 90-92 K was obtained reproducibly, while the $T_{CO}$ for the YBCO film grown by conventional PLD was only 84-86 K.

Typical 3-dimensional AFM images for the STO/YBCO bilayers are shown in Fig. 3, in which the thickness of the STO film is (a) 60 nm, (b) 30 nm, (c) 15 nm, and (d) 3 nm, respectively. We found that the grain size increased with the STO film thickness. Moreover, nonuniformity in thickness, exhibited along the interface of the STO/YBCO bilayers, made it hard to decide the effective thickness of the insulating STO layer, and also increased the difficulty of fabricating good tunneling junctions. However, no large protrusion due to the
The S/I/S junction suffers severely from the pinhole problem, which arises from droplets and particles, whenever conventional PLD is employed. By applying eclipse PLD for the YBCO/STO/YBCO heteroepitaxy, the pinhole problem was effectively eliminated. Typical tunneling conductance of our junction at $T = 4.9$ K was shown in Fig. 4. The arrows indicate the gap-like structures occur at bias around $9$ mV, $15$ mV, $25$ mV, and $40$ mV, respectively. The two main features of the tunneling characteristics of the high-$T_c$ S/I/S junctions, namely, the zero-bias anomaly and anisotropic gap $[8-11]$, are exhibited in Fig. 4. Our tunneling results resemble those of K. Hirata et al. $[9]$ and T. Kusumori et al. $[10]$ obtained for YBCO/Y$_2$O$_3$/YBCO and YBCO/CeO$_2$/YBCO, respectively. The observed gap structure did not resemble the typical BCS curve. We interpret this in the following way $[10]$. The spiral growth of a YBCO film $[12]$ allows the tunneling processes in both the ab-plane and the c-axis despite the c-axis orientation of the film used. Then tunneling processes between the c-axes, between the ab-planes, and between the c-axis and the ab-plane are expected. Because of the c-axis orientation of the films, the first dominant process may be the tunneling between the c-axes, the second one between the c-axis and the ab-plane, and the third one between the ab-planes. With the assumption that the anisotropic gap parameter $\Delta_{ab}$ and $A_r$ are equal in both electrodes, the structure at about $25$ mV may be due to the tunneling between the ab-plane and the c-axis, hence $\Delta_{ab} + A_r = 25$ meV. The structures at around $9$ mV, $15$ mV and $40$ mV may be associated with $2\Delta_{ab} = A_c$ and $2\Delta_{ab}$, respectively. The values $\Delta_{ab} = 19-21$ meV and $A_c = 4-5$ meV roughly satisfy the above requirements. The deduced YBCO gap values of our YBCO/STO/YBCO junctions are consistent with those of YBCO/CeO$_2$/YBCO $[10]$ and Pb/STO/YBCO $[13]$. 

![FIG. 4. Typical tunneling conductance for YBCO/STO/YBCO junction at T = 4.9 K. The arrows indicate the gap-like structures occur at bias around 9 mV, 15 mV, 25 mV, and 40 mV, respectively.](image)
IV. Conclusion

A shadow mask was introduced into the pulsed laser deposition of the YBCO films. Using this eclipse PLD, YBCO thin film with a droplet-free surface, a good superconducting and tunneling property has been obtained. The anisotropic gap values of YBCO deduced from our YBCO/STO/YBCO junctions are consistent with the existing reports.

References