Preparation of superconducting multilayer structure for power electronic devices

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Abstract

Multi-step KrF excimer pulsed laser deposition (MSPLD) technique has been developed to fabricate high-quality micrometer thick superconducting YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) films. As has been shown, a crystallization of YBCO at the initial stage of deposition crucially affects on thick YBCO film performance. Smooth, c-axis oriented 1-μm thick YBCO films with the critical temperature of $T_c$ (zero) = 89.5 K and the overall critical current density of $J_c$ (77 K) = 10$^3$ A/cm$^2$ have been prepared on the single crystal MgO substrate in three-step process by monitoring the laser fluence, laser repetition rate and substrate temperature. Micrometer thick YBCO films on the single crystal LaAlO$_3$ substrate show $T_c$ (zero) = 85.7 K and $J_c$ (77 K) = 10$^3$ A/cm$^2$, indicating that optimization of MSPLD processing conditions is much more critical for substrates with a small substrate film lattice mismatch. Furthermore, Pb(Zr,Sn)O$_3$/PZT/YBa$_2$Cu$_3$O$_{7-x}$/MgO heterostructures have been prepared by MSPLD technique with the critical temperature of $T_c$ (zero) = 82.0 K in superconducting YBCO layer. © 1998 Elsevier Science S.A.

Keywords: Superconducting multilayer structure; Power electronic devices; YBCO film

1. Introduction

High-$T_c$ superconducting HTSC thin films have drawn much attention due to their application in power electronic devices, such as current leads, current limiters and switches, as well as superconducting microcircuits for microwaves. To make HTSC films applicable for power electric devices, such as wires and fault current limiters, it is required to prepare thick films with high critical current density $J_c$ and large cross section area. However, in HTSC films with thicknesses over 400 nm, it is hard to obtain high $J_c$. Conventional deposition techniques that have been applied for thick film preparation do not achieve a perfect orientation across the total film thickness.

Also, to fabricate HTSC electronic devices, like superconducting field effect transistor (SuFET), high-quality heteroepitaxial multilayers of superconductors and insulators are required. Lin et al. [1] reported the SuFET device using PZT/YBCO heteroepitaxial multilayer as a gate layer. Although some important problems in the process and properties of superconductor/insulator and superconductor/ferroelectric multilayers are going to be overcome, superconducting critical parameters and dielectric characteristics must be still improved for its practical use in microelectronics.

Many groups have reported that some important characteristics of superconducting films are established by interactions on the film/substrate interface. Streiffer et al. [2] studied ultra-thin films of YBa$_2$Cu$_3$O$_{7-x}$ on MgO substrates. Zheng et al. [3] have used scanning tunnelling microscopy (STM) to study YBCO epitaxially grown on MgO and SrTiO$_3$. Alarco et al. [4] examined the early stages of growth of YBCO on YSZ substrate using STM. They inferred to improve superconducting and microstructural properties of layer by layer grown films, multilayered thick films and heterostructures. One has to take care of processing conditions at the initial stage of deposition to obtain epitaxial growth of crystallines on the interfaces. However, superconducting film properties of deposited multilayer structures have not been reported.

In this study, high quality micrometer thick YBCO films have been prepared using multi-step pulsed laser deposition technique (MSPLD). Films show high c-axis orientation, smooth surface morphology, $T_c$ (zero) = 89.5 K and overall critical current density of $J_c$ (77 K) = 10$^3$ A/cm$^2$. Furthermore, PZT/YBCO heteroepitaxial multilayers have been fabricated. They exhibit superconducting...
transition at $T_c(\text{zero}) = 82.0$ K and look promising to be used in high power device applications.

2. Experimental setup

A pulsed KrF excimer laser (Lambda Physik LPX-305icc, wavelength $\lambda = 248$ nm, pulse duration = 25 ns, maximum output = 850 mJ) was used to ablate the superconducting YBa$_2$Cu$_3$O$_{7-x}$ ceramic target. The laser beam was focused on the target through a lens. The spot size of the beam was $0.5 \times 20$ mm$^2$ on the target with an incident angle of 45°. A substrate was placed at a distance of 50 mm from the target. A deposition chamber was exhausted up to the base pressure of $10^{-4}$ Pa by a rotary and a

Fig. 1. SEM micrographs of 1-µm thick YBCO films deposited at different initial laser fluence and same laser repetition rate of 1 Hz and MgO substrate temperature $T_s = 710^\circ C$. 
turbo-molecular pumps. The substrate was heated up to 600–710°C using the infrared lamp. Pure oxygen gas was introduced into the chamber and total deposition pressure was maintained at 29 Pa (200 mTorr). After the deposition, oxygen gas was introduced into the deposition chamber up to atmospheric pressure and films were cooled down to 50°C (in-situ annealing).

For the PZT layer deposition, sintered Pb(Zr0.52Ti0.48)O3 tablet has been used as a target. The substrate temperature, total deposition pressure of oxygen, laser repetition rate and laser energy density were as follow: 650°C, 15 Pa (100 mTorr), 10 Hz and 2 J/cm², respectively.

The surface morphology, microstructure and superconducting properties of these films were investigated by scanning electron microscopy, X-ray diffraction and DC four probe method, respectively.

3. Results and discussion

3.1. YBa2Cu3O7−x thick films on MgO(100) substrates

YBCO thick films on MgO substrate using MSPLD technique were studied. At first we attempted the two step PLD process. We selected different deposition conditions to grow initial ‘template’ YBCO layer (100 shots): substrate temperature (690–710°C), laser fluence (0.5–5 J/cm²) and laser repetition rate (1–20 Hz). Processing conditions of the following deposition (30,000 shots) were kept constant to be optimum for preparation of YBCO thin film. Substrate temperature, laser fluence and repetition rate were as follow: 710°C, 2 J/cm² and 10 Hz, respectively. Surface morphology and crystal orientation have been found strongly dependent on deposition conditions of initial growth. Fig. 1 shows SEM photograph of film surface. At the lower laser fluence (Fig. 1a), large grains are observed on the film surface and crystalization is deficient due to insufficient migration energy at the substrate surface. Besides the higher laser fluence (Fig. 1c), large spherical particulates have been found on the film surface and deposited film was not crystallized. It happens because large droplets being ejected from the target surface and having large kinetic energy bombad deposited film and destroy its crystallinity. Furthermore, if initial laser repetition rate was high, surface morphology and crystal growth degraded because nucleation of YBCO phase occurs in thermodynamically non-equilibrium conditions. The optimum deposition conditions for MgO(100) substrate interface have been found as follow: substrate temperature of 710°C, laser fluence of 3 J/cm² and laser repetition rate of 1 Hz. However, the superconducting properties of prepared thick film have been found much worse than thin YBCO films made by conventional PLD technique: \( T_c(zero) = 82.1 \text{ K} \) and \( J_c(77 \text{ K}) = 10^4 \text{ A/cm}^2 \). This is an agreement with many studies which show that superconducting properties degraded when film thickness exceeds the value of 400–500 nm [5].

Therefore, to improve the superconducting properties of thick films, we applied three-step deposition conditions process shown in Fig. 2. Deposition conditions for initial growth (100 shots) of template YBCO layer were fixed: substrate temperature of 710°C, laser fluence of 3 J/cm² and laser repetition rate of 1 Hz. At intermediate stage of deposition (10,000 shots) substrate temperature, laser fluence and repetition rate were kept at 710°C, 2 J/cm², 10 Hz, respectively. Deposition conditions for the third deposited layer (20,000 shots) were as follow: substrate temperature (650–710°C), laser fluence (0.5–5 J/cm²) and laser repetition rate (5–20 Hz). After deposition, oxygen gas was introduced into the deposition chamber up to atmospheric pressure and film was cooled down to 50°C. Optimum deposition conditions are listed in Table 1.

The best YBCO thick films with high-\( T_c \), smooth surface morphology and low resistivity at room temperature were obtained at lower laser repetition rate as 5 Hz and substrate temperature is 710°C at the final third stage of deposition. If substrate temperature at the final stage of deposition is lower, \( a \)-axis orientation appears. Therefore, to obtain highly crystalline thick YBCO/MgO films, laser repetition rate showed be lowered to adjust the crystallization energy at the final stage of deposition. The optimum deposition condition for the final stage has been found as follow: laser repetition rate of 5 Hz, laser energy density of 2 J/cm² and substrate temperature of 710°C.

3.2. YBa2Cu3O7−x thick films on LaAlO3 substrates

LaAlO3 substrate is one of the most attractive substrate material because of good lattice match with YBCO, low dielectric constant and low loss tangent. YBCO thin films on LaAlO3 show good superconducting properties and reproducibility. The multi-step deposition procedure was utilized for YBCO thick films onto LaAlO3 substrate. Deposition procedure was the same as film preparation on MgO substrate.

When we change the initial growth conditions by the template YBCO layer, film properties were not quite dif-

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Fig. 2. Schematic view of multilayered film made by multistep pulsed laser deposition.
Table 1
Optimum deposition conditions for YBCO thick films

<table>
<thead>
<tr>
<th>Substrate temperature</th>
<th>710°C</th>
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<tbody>
<tr>
<td>O₂ pressure</td>
<td>200 mTorr</td>
</tr>
<tr>
<td>Substrate</td>
<td>MgO(100) LaAlO₃(100)</td>
</tr>
<tr>
<td>Lattice mismatch YBCO</td>
<td>8%</td>
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<tr>
<td>Initial deposition layer (300 shots)</td>
<td>laser repetition rate</td>
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<td></td>
<td>laser fluence</td>
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<tr>
<td>Intermediate deposition layer (10,000 shots)</td>
<td>laser repetition rate</td>
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<td></td>
<td>laser fluence</td>
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<tr>
<td>Final deposition layer (20,000 shots)</td>
<td>laser repetition rate</td>
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<tr>
<td></td>
<td>laser fluence</td>
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<tr>
<td>Tₚ(zero)</td>
<td>88.2 K</td>
</tr>
<tr>
<td>Tₚ(onset)</td>
<td>91.5 K</td>
</tr>
<tr>
<td>Overall critical current density Jc (at 77.3 K)</td>
<td>10^3 A/cm²</td>
</tr>
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different. Because lattice mismatch between YBa₂Cu₃O₇₋ₓ and LaAlO₃ is rather small, it is easy for species to migrate along the substrate surface. Furthermore, optimum deposition conditions for intermediate and final deposited layers have been found much wider than the optimum conditions for MgO substrate. It suggests easiness to reproduce YBCO films with the same quality. The optimum conditions for each layer were the following: laser repetition rate of 5–10 Hz, laser energy density of 3–4 J/cm² and substrate temperature of 710°C. Changing of the conditions for multilayers deposition was not effected. Fig. 3 shows X-ray diffraction patterns of YBCO thick films on MgO and LaAlO₃ substrates. YBCO films on the MgO substrate show preferential c-axis orientation (Fig. 3a). Although it is using LaAlO₃ substrate, a-axis orientation has been revealed (Fig. 3b). Furthermore, many grain domains have been observed in SEM photographs on the surface of 1 µm YBCO film made on LaAlO₃ substrate by MSPLD. Their surface morphology is much rougher than that of YBCO thick film onto MgO substrate and is the same as prepared by the usual sequence of deposition conditions.

Fig. 4 shows resistive transition in YBa₂Cu₃O₇₋ₓ thick film on LaAlO₃ substrate made by MSPLD. In samples prepared not using multi-step deposition, it is hard to obtain superconducting transition above 77 K. The sample shown in Fig. 4b has Tₚ(zero) = 85.7 K and Tₚ(onset) = 91.3 K. The normal resistivity is higher and the critical current density of 10^3 A/cm² is lower than for YBCO thick films prepared on MgO substrate. It suggests that optimization of MSPLD processing conditions for LaAlO₃ substrate is difficult due to a small substrate/film lattice mismatch.

3.3. Pb(Zr,Ti)O₃ / YBa₂Cu₃O₇₋ₓ / MgO(100) heterostructures

We also prepared PZT/YBCO heterostructure on MgO substrate. Thickness of YBCO and PZT films was 500 nm and 300 nm, respectively. The YBCO layer was deposited using multi-step deposition technique. The layer-by-layer SEM analysis indicated that surface morphology of PZT is strongly affected by smoothness of YBCO layer.

X-ray diffraction spectra show c-axis orientation for PZT film indicating PZT films onto YBCO films have good crystallinity.

Fig. 5 shows temperature dependence of resistivity for PZT (300 nm)/YBCO (500 nm) heterostructure. This multilayer shows high normal resistivity and Tₚ(zero) of 82.0 K.
Fig. 5. Superconducting transition of the YBCO layer in the PZT(300 nm)/YBCO(500 nm)/MgO heterostructure.

It is shown that preparation of the smooth surface of YBCO bottom layer is dominant to obtain high quality PZT/YBCO heterostructures.

4. Conclusion

YBa$_2$Cu$_3$O$_{7-x}$ multilayer structures for power electronics devices have been prepared by novel multi-step KrF excimer pulsed laser deposition technique. Surface roughness of these films is strongly dependent on the deposition conditions for the first template layer as well as substrate material. The most crucial issue is to control film/substrate interface interaction for the first template oxide layer. To obtain high quality heterostructures, it is important to take care of surface roughness of deposited films. Multi-step pulse laser deposition technique could be easily applied to fabricate different complex oxide heterostructures.

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References