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FABRICATION OF HIGH QUALITY $YBa_2Cu_3O_y$ THIN FILMS USING PULSED LASER DEPOSITION

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ABSTRACT

High quality $YBa_2Cu_3O_y$ (YBCO) thin films for directly coupled dc superconducting quantum interference device (SQUID) were fabricated by pulsed laser deposition. Several critical parameters have been optimized through systematic studies. Thus, the films showing the T_C of above 91K and J_C of above $2\times10^6 A/cm^2$ at 77K were routinely obtained. Extensive AFM and X-ray diffraction studies have been conducted for morphological and structural analyses. The directly coupled DC-SQUIDs were fabricated from the YBCO thin films deposited on $SrTiO_3$ bicrystals under the optimized conditions. The measurement on $2I_C$ and swing voltage give $200\mu A$ and $17\mu V$ at 77K, respectively.

INTRODUCTION

Pulsed laser deposition (PLD) has become a popular method for fabricating many types of thin films. Especially in the field of high-T_c superconducting thin films technologies, PLD is one of the most preferred methods because high quality thin films of complicated compositions could be obtained easily at relatively low cost. Furthermore, the PLD technique is advantageous over other existing techniques due to its high deposition rate and the ease of multi-layer stacking in-situ. When fabricating a device, the quality of thin film such as surface morphology and uniformity is as important as electrical properties. For the past few years, there have been many efforts to improve film properties such crystal orientation as in-plane alignment in addition to other electrical properties.

In this article, we systematically studied the effects of several processing parameters on electrical properties, crystal orientation and surface morphology of YBCO thin films using PLD. To confirm the film quality we have measured the device characteristics of directly coupled dc SQUID magnetometer using the above films.

EXPERIMENTAL

A 248-nm KrF excimer laser (pulse duration=25ns, Lambda Physik, LPX 305i) operating at 5 Hz was used for the film deposition. The laser beam was focused through a lens to about 1×4.5㎟ size onto a stoichiometric YBCO target with an energy density of about 1 J/㎠. Substrates, mounted on the heater (US Thin Film Pro. Inc.) by silver paste, were placed in front of the target at the distance of 40~80㎜. Substrate temperature (T_s) during deposition was varied from 700 to 840℃ and the ambient O_2 pressure (PO_2) was applied in the range of 100 to 700 mTorr. YBCO films of 100~200 nm thickness were deposited onto 10×10㎟ MgO and $LaAlO_3$ substrates of (100) orientation. For the bicrystal grain boundary junction SQUID magnetometer, we used (100) $SrTiO_3$ bicrystal with a 24° miso-rientation angle. The target pellet was rotated during the irradiation to minimize needle-like morphology on its surface[1]. Immediately after the deposition, pure oxygen was introduced into the deposition chamber and T_s was decreased to 550℃ at a rate of 15℃/min. After annealing at this temperature for 45min, Ts was cooled down to the room temperature. The films were characterized by x-ray diffraction (both θ-2θ and in-plane scans), AFM (Atomic Force Microscopy) and micro-Raman spectroscopy. The superconducting transition temperatures (T_C) and critical current densities (J_c) of YBCO films were resistively measured by a standard four-probe geometry on 30-㎛-long and 5-㎛-wide bridges.

RESULTS AND DISCUSSION

Figure 1 shows the effect of the T_s during deposition on the T_cs of the YBCO thin films grown on MgO and $LaAlO_3$ substrates. All of the films represented here were deposited under 500mTorr oxygen and the target-substrate distance was 60㎜. When MgO substrates were used, T_C was increased gradually with T_s of above 720℃ showing maximum T_C of 91.5K at 800℃. Beyond 800℃, T_C decrea-sed abruptly. In case of $LaAlO_3$ substrates, similar increase of T_C was observed, but temperature range, showing Tcs of about 90K, was much wider than MgO substrates. Over a range of T_s of 100℃ (740 to 840℃) the films on $LaAlO_3$ showed T_cs of above 89K. Depositing films above 840℃ was not available due to heating system limitations.

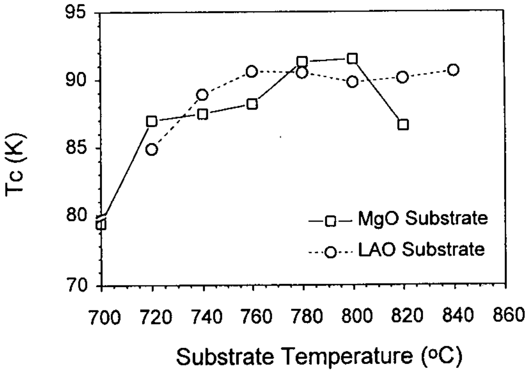

Fig. 1. The effect of the T_s during deposition on the T_cs of the YBCO thin films made by PLD

From the θ-2θ scan data, it was observed that when T_s was above 760℃, all the films represented here were oriented with its c-axis perpendicular to the substrate surface regardless of the substrates used.

Figure 2 shows PO_2 dependences of the T_cs on MgO and $LaAlO_3$ substrates. As shown in Fig. 2, the PO_2 during the deposition also had a substantial effect on the Tcs of films. T_s for the films, represented here, on MgO and $LaAlO_3$ were at 800 and 780℃, respectively. The other parameters were the same as those

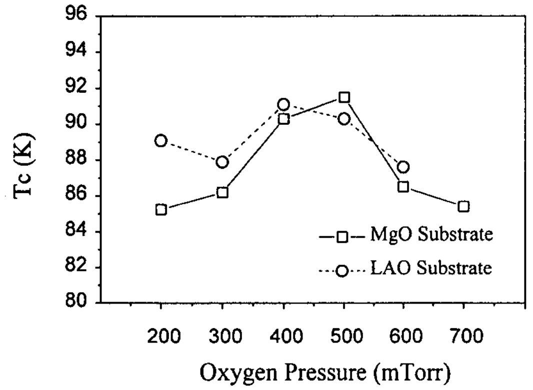

Fig. 2. The effect of the PO_2 during deposition on the T_cs of the YBCO thin films made by PLD

of Fig. 1. The films grown under the PO_2 of 400~500mTorr showed the best T_C of above 90K, irrespective of the substrates used. Both higher and lower PO_2 resulted in lower T_C.

For the growth of YBCO thin films, it is highly recommended to maintain the deposition chamber under an oxidizing environment to compensate for some loss of a constituent oxygen. This is why the deposited films tend to be deficient in oxygen when the ablation is done in vacuum. Other roles of background oxygen is to provide help for the formation and stabilization of the desired crystal phase at the deposition temperature[2]. In-situ processing of YBCO films typically requires 100~300mTorr of the PO_2 in the chamber. 400~500mTorr of the PO_2, as represented here, is somewhat higher. Higher in PO_2 is thought to be due to relatively higher Ts when compared with others (~750℃). This correlation could be obtained from the stability phase diagram of YBCO as a function of temperature and oxygen pressure[3].

While the surface morphology of the film was also influenced by PO_2, it was confirmed, from AFM analysis, that the films grown above 500mTorr contain many small-sized particulates, so called boulders, ranging 0.1 to 0.5㎛, on the surface. It is noted that the optimum PO_2 value range is 400~500mTorr (Fig. 2).

Figure 3 shows the effect of target to substrate distance on the T_cs of YBCO films on MgO. All the processing parameters are the same as that of Fig. 2, except keeping PO_2 of 400mTorr fixed. The target to substrate distance, showing T_C above 89K, was in the range of 50~60㎜. As reported earlier by several groups, the velocities of the species in the laser plume are more than 10^6cm/s near the target, and are decelerated as a function of ejected distance due to the collision with O_2[4]. These energetic species may cause some damage to the film if the target-substrate distance is too close. On the other hand, if the substrate is too far away, the deposition rate will be too low and the kinetic energy of the ablated species will be significantly lowered by the collision with surrounding gases as well as each other. This leads to lowering the surface diffusivity of deposited species and results in the poor film quality.

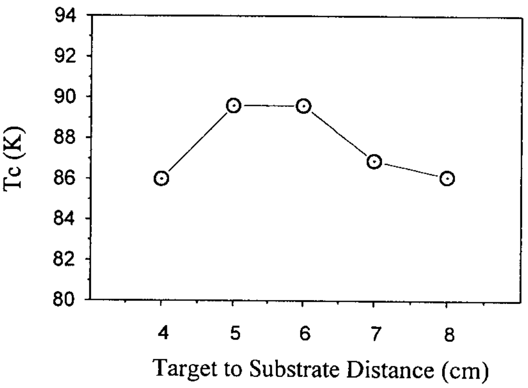

Fig. 3. The effect of target-substrate distance on the TCs of the YBCO thin films made by PLD

Therefore, the substrate should be placed in the optimum distance range from the target to produce a good quality film. It is worthwhile to note that the optimum distance, under any processing parameters, is comparable to plume size. The optimum target-substrate distance of 50~60㎜, as shown Fig. 3, is exactly the plume length.

Figure 4 shows the resistance as a function of temperature for YBCO films deposited on $LaAlO_3$ under the optimized processing parameters. The detailed parameters are shown in inset in Fig. 4. For $T>T_C$, the resistance behavior is metallic and can be extrapolated to the origin, as shown in Fig. 4. The zero resistance temperature ($T_{c,\,o}$) as high as 90 ~92K was obtained routinely. Similar results were obtained in the case of MgO substrates ($T_{c,\,o}=$~91K).

Figure 5 shows a θ-2θ x-ray diffraction (XRD) pattern for a YBCO film deposited on MgO at 800℃ at the O_2 pressure of 500 mTorr. One can see only (00l) peaks, indicating that the film is c-axis textured.

Figure 6 shows the x-ray φ-scan taken at (104) reflection of a YBCO film on MgO to determine in-plane epitaxy. The 0°~360° scan shows only four peaks, meaning that the film made with the optimized processing parameters has an excellent in-plane epitaxy . This in-plane epitaxy was also confirmed by a polar plot analysis[5].

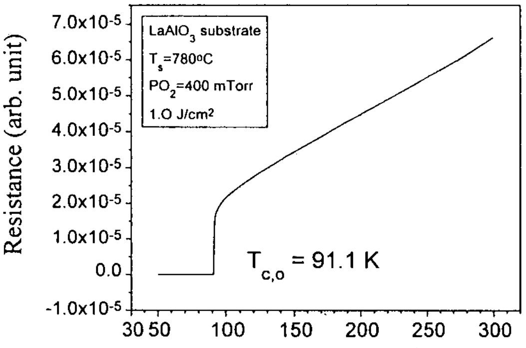

Fig. 4. Resistance vs temperature characteristic of a YBCO thin film grown by PLD on $LaAlO_3$ substrate

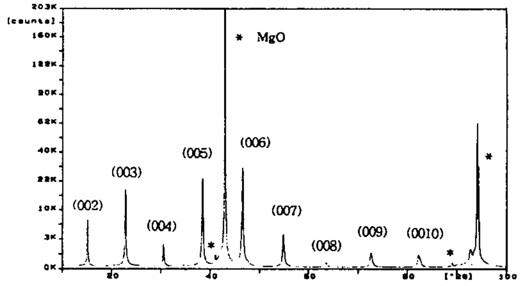

Fig. 5. θ-2θ x-ray diffraction (XRD) pattern for a YBCO film deposited on MgO at 800℃ under the O_2 pressure of 500mTorr.

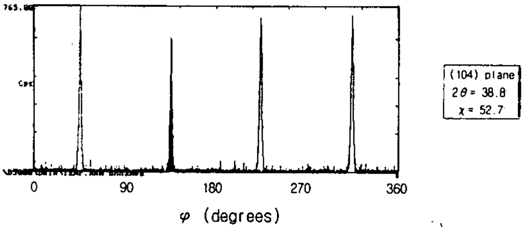

Fig. 6. x-ray phi scan for a YBCO thin film grown by PLD on MgO substrate.

Figure 7 shows a plot of J_C for a YBCO film on MgO as a function of temperature. At 77 K and zero field, the J_C was found to be 6×106 A/㎠. This high valve of JC could be obtained partly due to the in-plane epitaxy in the YBCO layer on the substrates, as confirmed in Fig. 6.

Figure 8 shows a typical AFM picture of a YBCO film on MgO deposited at 500mTorr PO_2 and Ts of 800℃. Several particulates with sizes of 0.2~1.0㎛ could be seen over the whole 20×20㎛² scan area. While the ori-

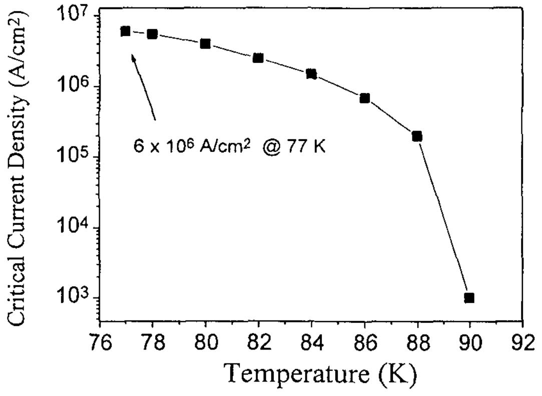

Fig. 7. Critical current density (Jc) measurement for a YBCO thin film deposited on MgO substrate.

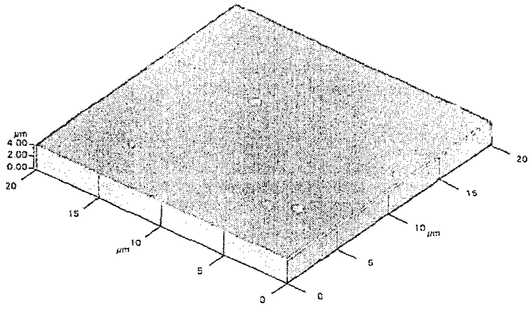

Fig. 8. AFM image of a YBCO thin film on MgO deposited at 500mTorr PO_2 and T_S of 800 ℃.

gin of the particulates is still under discussion, it is generally known that the irregular-shaped particles larger than about submicron are directly transferred from the target to the substrate. On the other hand, the sizes of the particles formed from the vapour phase tend to be in the nanometer range[6]. Considering the size and shape of the particulates formed on the surface, these are thought to be introduced directly from the target.

One of the applications of our high-quality thin films using PLD is the fabrication of the SQUID magnetometers[7]. We hasve made directly coupled de SQUID magnetometers from single layers of YBCO on $SrTiO_3$ bicrystal substrates[8]. At 77K, the SQUIDs showed RSJ (Resistively Shunted Junction) like current-voltage (I-V) characteristics and voltage modulation in response to external fields.

Figure 9 shows the current-voltage (I-V) characteristic of a dc SQUID magnetometer at 77K. The junctions have critical current, 2IC, of 200μA and normal state resistance RN of 0.5Ω at 77K. Voltage-flux (V-Φ) mea-surement showed the maximum peak to peak modulation voltage of 17μV (Fig. 10).

Figure 11 shows a rms flux noise of the SQUID at 77K in zero field, obtained using flux-locked loop electronics. The noise characteristics were measured in magnetically shielded box. The white noise of the magnetometer is $13\mu\Phi_0/\sqrt{Hz}$ at 400Hz, rising to $60\mu\Phi_0/\sqrt{Hz}$ at 10Hz. These flux noise values correspond to noise energy 7.24×10^{-30} J-sec and 1.54×10^{-28} J-sec, respectively.

These obtained values are low enough to measure the biomagnetic signal from the heart of body.

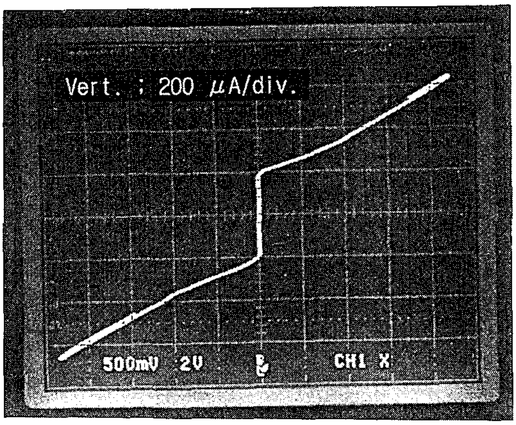

Fig. 9. I-V characteristic of a YBCO thin film grown by PLD on $SrTiO_3$ bicrystal substrate.

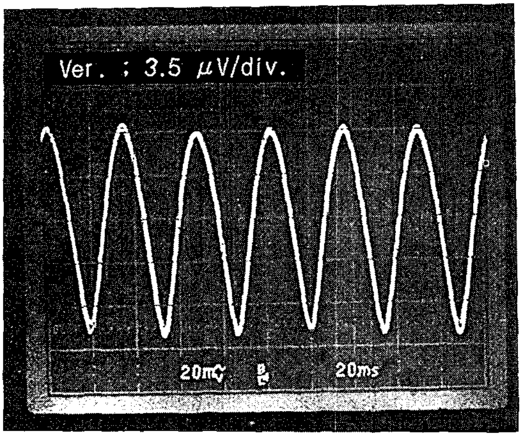

Fig. 10. V-Φ characteristic of a YBCO thin film grown on $SrTiO_3$ bicrystal substrate.

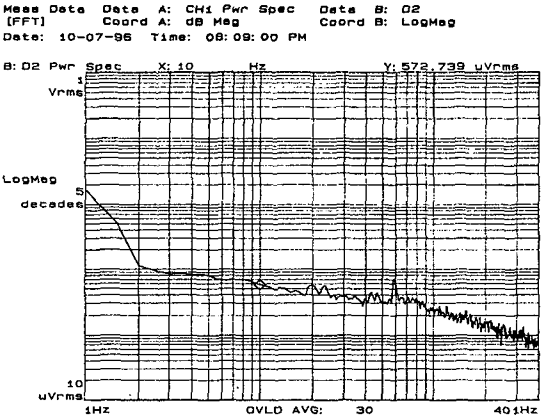

Fig. 11. Measured rms flux noise, $\sqrt{S\Phi(f)}$, of the dc SQUID made by the YBCO thin film grown on $SrTiO_3$ bicrystal substrate.

SUMMARY

Critical processing parameters of PLD for high-quality YBCO thin films on MgO and $LaAlO_3$ substrates have been optimized through systematic studies. YBCO thin films on MgO and $LaAlO_3$ showing the best characteristics could be made under 500 mTorr oxygen at 800℃ and 400 mTorr at 780℃, respectively. Considering the plume lengths formed under respective parameters, target-substrate distance of 50~60㎜ is found to be adequate for obtaining high-quality films. Using obtained parameters to date, the films having T_C of above 91K and J_C of above 2×10^6 A/㎠ at 77K have been obtained reproducibly. Extensive X-ray diffraction studies suggest that the YBCO film shows excellent in-plane epitaxy as well as its c-axis orientation perpendicular to the substrate surface. The directly coupled DC-SQUIDs with bicrystal grain boundary junction on $SrTiO_3$ substrates using above thin films show 2IC and swing voltage of 200μA and 17μV at 77K, respectively. The flux noise of the magnetometer is $13\mu\Phi_0/\sqrt{Hz}$ at 400Hz and $60\mu\Phi_0/\sqrt{Hz}$ at 10Hz.

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