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PREPARATION OF $YBa_2Cu_3O_{7-\delta}$ EPITAXIAL THIN FILMS BY PULSED ION-BEAM EVAPORATION

S. Sorasit, G. Yoshida, T. Suzuki, H. Suematsu, W. Jiang and K. Yatsui

Extreme Energy-Density Research Institute, Nagaoka University of Technology
Nagaoka, Niigata 940-2188, Japan

ABSTRACT

Thin films of $YBa_2Cu_3O_{7-\delta}$ (Y-123) grown epitaxially have been successfully deposited by ion-beam evaporation (IBE). The c-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin films were successfully deposited on MgO and $SrTiO_3$ substrates. The Y-123 thin films which were prepared on the $SrTiO_3$ substrates were confirmed to be epitaxially grown, by X-ray diffraction analysis. The instantaneous deposition rate of the Y-123 thin films was estimated as high as 4 mm/s.

I. Introduction

After the discovery of the first high T_c superconductor, $La_2Cu_4O_{4+\delta}$ [1], many superconductors have been found. $YBa_2Cu_3O_{7-\delta}$ (Y-123) is one of the superconductors and has a peak in the characteristics of critical current density (J_c) as a function of applied magnetic field (H) [2]. This phenomenon is called the peak effect. Many works have been reported on the peak effect in Y-123. The reason of the peak effect, however, has not been clarified.

In order to study the peak effect, Y-123 samples which are epitaxially grown thin films with thickness of ~ 10 μm are required. Numerous thin films preparation techniques, such as sputtering [3], molecular beam epitaxy (MBE) [4], electron beam evaporation [5] and pulsed laser ablation methods [6] have been developed to prepare the epitaxailly grown Y-123 thin films. However, the deposition rate of Y-123 thin films by these methods was slow and it is difficult to prepare 10 μm thick films.

Ion-beam evaporation (IBE) method has been developed to prepare various kinds of thin films with high deposition rates. The preparation of the films was carried out with a intense pulsed ion beam generator, which produces an ion beam with a kinetic energy of 1 MV (peak) and a current of 60 kA for a pulse width of ~50 ns [7]. Then the ion beam is irradiated on a target. Since the *range* of the ion beam in the target is very short, the surface is heated according to the delivered energy density by the ion beam. This causes the production of high-density ablation plasma which deposits on substrates with a rate of

~1μm / shot [8].

In the present work, the Y-Ba-Cu-O thin films are prepared by the ion beam evaporation (IBE) method. The crystallinity of the Y-123 thin films is characterized by various X-ray diffractometries.

II. Substrate Materials

Single crystals of MgO are known as substrate materials which do not react with Y-123 thin films at growth temperatures [9]. The preparation of high quality c -axis oriented Y-123 thin films have been reported on the MgO substrates [10], despite more than 9 % lattice mismatch between a-parameters of Y-123 and MgO.

Lattice mismatch between $SrTiO_3$ and Y-123 (1%) is better than that of Y-123 and MgO. Utilizing this advantage, epitaxially grown Y-123 thin films were prepared on $SrTiO_3$ substrates. Moreover, the thermal expansion coefficient of $SrTiO_3$ is close to that of Y-123. From these reasons, single crystals of MgO and $SrTiO_3$ were selected as substrates on which Y-123 thin films were deposited by IBE.

III. Experimental Arrangement

Figure 1 shows the schematic of the ion beam source and the sample deposition chambers in the pulsed power generator "ETIGO-II". The ion beam was produced by a magnetically insulated ion diode (MID) with geometrically focused configuration. A polyethylene sheet (flashboard) was attached on the anode (aluminum). The ion species were mainly protons. The cathode, which also worked as a one-turn theta-pinch coil, produced a transverse magnetic field, by which electrons emitted from the cathode are prevented from reaching the anode. The obtained ion beam was focused on the sintered Y-Ba-Cu-O target. After the ion beam irradiation, ablation plasma is produced on the surface of the target and thin films were deposited on the single crystal substrates facing toward the target. For each substrate, thin film was deposited by a single shot of ion beam

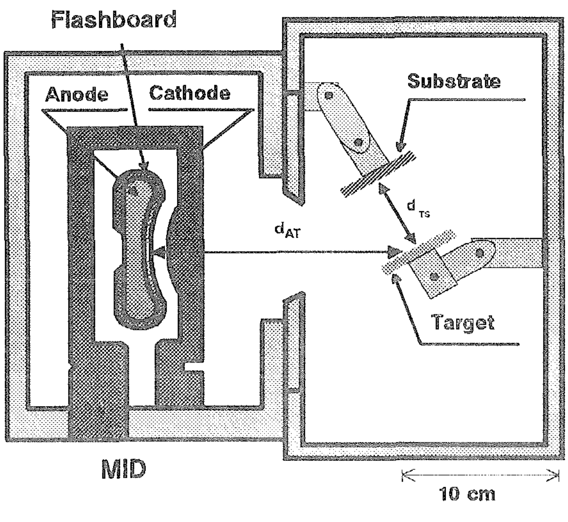

Fig. 1 Schematic of the ion beam source and the sample deposition chambers.

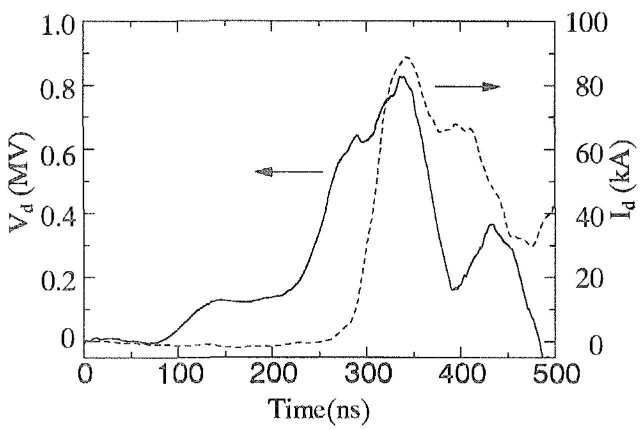

Fig. 2 Typical waveforms of V_d and I_d.

bombardment. Table I summarizes the deposition conditions.

Table I. Experiment conditions.

Accelerating Voltage	1 MV
Pulse width	50 ns
Ion type	H^+
Fluence	25 J/cm^2
Target	$YBa_2Cu_3O_{7-\delta}$
Target size	ϕ 50 mm, 5^t
Substrate	$SrTiO_3$ and MgO (100)
Distance (Anode- Target: d_{AT})	200 mm
Distance (Target-Substrate: d_{TS})	70 mm
Substrate temperature	RT.
Shot number	1 shot
Pressure	10^{-4} Torr

Table II. Annealing conditions.

900 ℃	5 hours
600 ℃	5 hours
400 ℃	40 hours

*These processes have been done in flowing oxygen gas

Figure 2 shows typical waveforms of V_d (diode voltage) and I_d (diode current). From Fig. 2, we see that V_d ~1 MV and I_d ~80 kA (peak) with τ (pulse width) ~ 70 ns (FWHM). Figure 3 shows the evolution of the ablation plasma which is formed on the Y-Ba-Cu-O target. These photographs were taken by a high-speed camera (Ultra NAC FS501). From these photographs we see that the Y-123 thin film was deposited in a period of 72 μs.

The as-deposited films were annealed in flowing oxygen gas. The annealing conditions are shown in Table II. The thin films were characterized with an X-ray diffractometer. The thickness of the thin films was measured by a roughness tester (Surfcom 130A, Tokyo Seimitsu). The cation composition of the target and the Y-123 grains in the thin films was determined by X-ray fluorescence spectrometry and energy dispersive X-ray analysis (EDX), respectively. A specimen for EDX was prepared by scratching the surface of a thin film on a $SrTiO_3$ substrate by a piece of glass. The powders from the film were collected on a carbon microgrid supported by a molybdenum mesh. The EDX spectra for the thin films was determined with an energy dispersive X-ray spectrometer attached on a transmission electron microscope (JEM-2010, JEOL) operated at 200 kV. The intensity of the characteristic X-ray from the specimen was calibrated by those from a standard sample of Y-123 powder. Thin film approximation which ignored absorption of the emitted X-ray in the films was assumed for quantifying the composition.

IV. Results and Discussions

The XRD patterns for the annealed Y-123 thin films on the MgO and $SrTiO_3$ substrates are shown in Fig. 4. The peaks of Y-123 thin films on both substrates exactly correspond

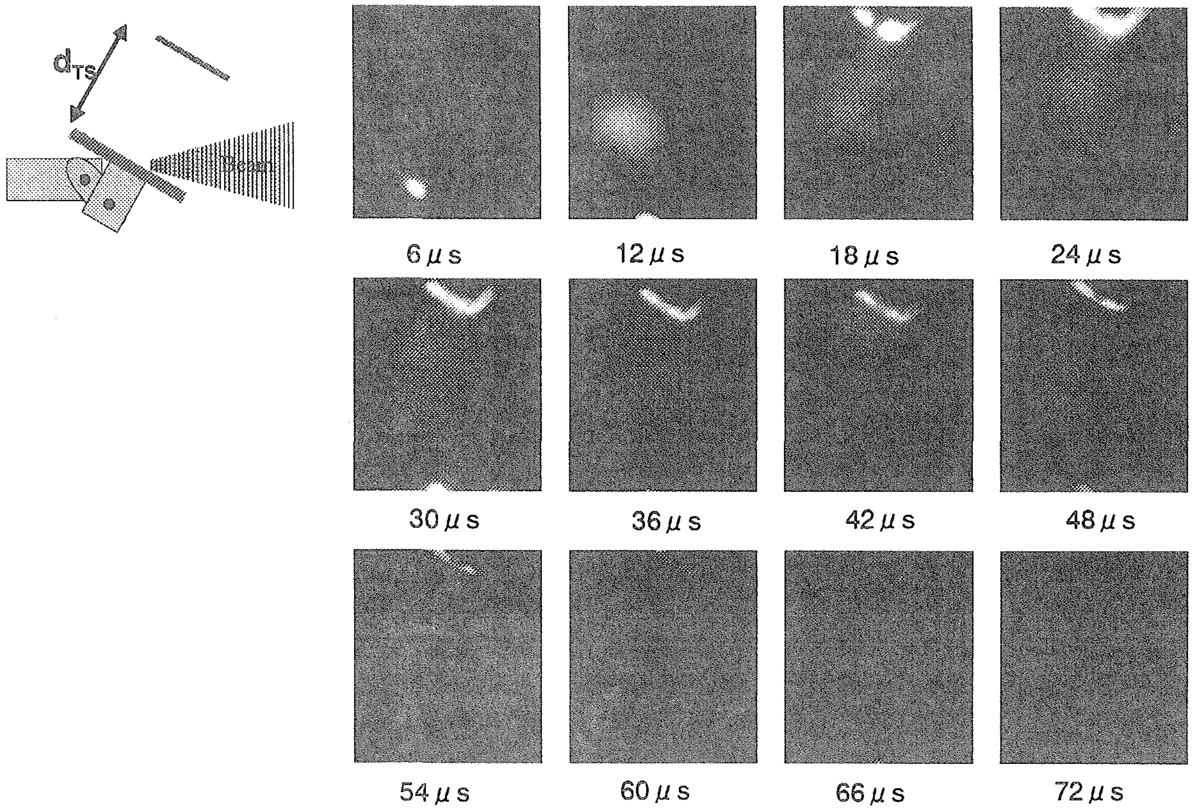

Fig. 3 High-speed photographs of the ablation plasma from the Y-Ba-Cu-O target.

to the reflections for Y-123 or substrates phases. The films were essentially single-phase. Although the peak for the 103 reflection is also found in the pattern for the Y-123 thin films on the MgO substrate, most of the peaks for Y-123 are matched to the 00 l reflections. This indicates that the c-axis for the Y-123 thin films was perpendicular to the (100) surface of the substrates.

The rocking curve of the 002 reflection has been taken to characterize the c-axis orientation. The full-width of half maximum (FWHM) value of 002 rocking curve are given in the Table III. From the results, we see that the Y-123 thin films on both substrates are less than 1° . This indicates that Y-123 thin films prepared by IBE method were c-axis oriented. Then, the intensity change of the (102) reflection for Y-123 thin films due to ϕ scanning have been taken to characterize the in-plane orientation, which are shown in Fig. 5. Since four-fold symmetry is clearly seen in the pattern for the thin films on the $SrTiO_3$ substrate, but not seen in the pattern for the Y-123 thin films on the MgO substrate, the Y-123 thin films are concluded to be epitaxially grown on the $SrTiO_3$ substrate.

The cation composition of Y-123 grains which was measured by EDX is shown in Table IV. From the result, the cation composition of the Y-123 thin films is almost the same as that of the Y-Ba-Cu-O target. This confirms the previously concluded character of the IBE method [11], i.e. the composition of the films is as same as that of target.

The thickness of the epitaxially grown Y-123 thin films was measured with the roughness tester, yielding 0.3 μm with a single shot of ion beam bombardment. According to the ablation plasma photographs which was shown in Fig. 2, the Y-123 thin films were deposited in a period of 72 μs. Thus, the instantaneous deposition rate of the films was calculated to be ~ 4 mm/s.

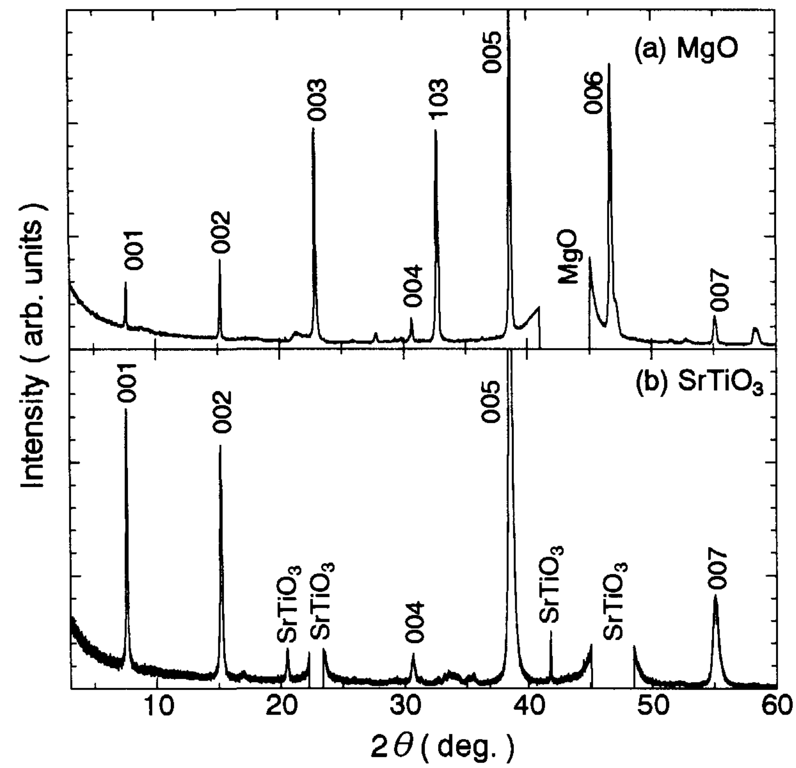

Figure 4 XRD patterns of the Y-123 thin films on (a) MgO and (b) $SrTiO_3$.

Table III. FWHM values of rocking curves of 002 reflections for Y-123 thin films.

Substrate	FWHM (deg.)
MgO	0.18
$SrTiO_3$	0.73

Table IV. Cation composition of the target and Y-123 thin film.

	Y	Ba	Cu
Target	1.0	2.0	3.0
Sample	1.0	2.0	2.9

V. Conclusions

The c-axis oriented Y-123 thin films have been deposited on the MgO and $SrTiO_3$. The epitaxial Y-123 thin films were successfully achieved on the $SrTiO_3$ substrate by IBE method. The composition of the films is found to be in good agreement with that of the target. Utilizing the high-density ablation plasma which was formed by this method, Y-123 thin films have been prepared by the fast deposition rate of ~ 4 mm/s.

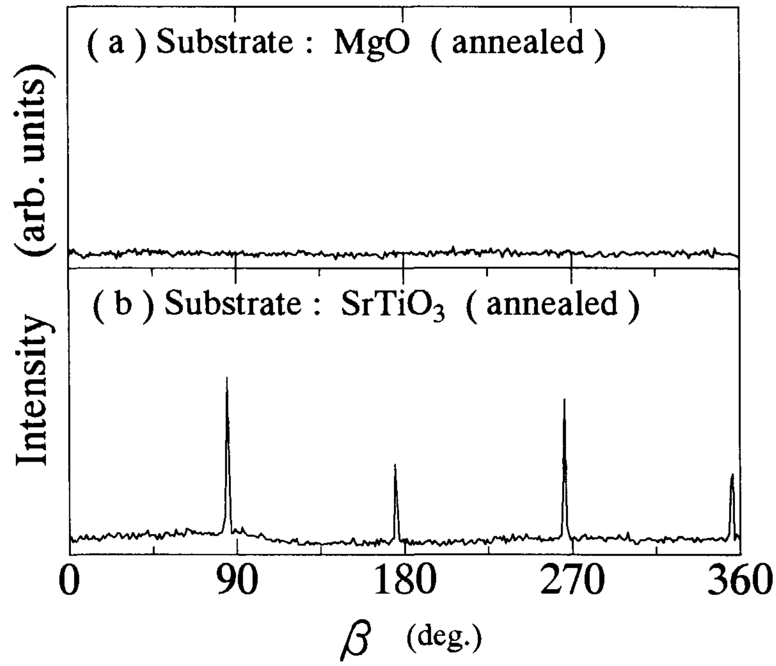

Figure 5 Intensity change of the (102) reflection due to β scan for Y-123 on (a) MgO and (b) $SrTiO_3$.

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