Crystal growth modified by pulsed laser irradiation on growing surface in Ba-Cu-O film preparation by laser ablation

CRYSTAL GROWTH MODIFIED BY PULSED LASER IRRADIATION ON GROWING SURFACE IN Ba-Y-Cu-O FILM PREPARATION BY LASER ABLATION

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ABSTRACT

$Ba_2YCu_3O_x$ superconducting films were prepared by Nd:YAG laser ablation, equipped with a second (ArF excimer) laser for irradiation onto the growing film surface. The irradiation onto the film during the deposition were delayed for various delay times against the ablation of $Ba_2YCu_3O_x$ target. The experiment showed that the second laser irradiation within several tens μs around the ablation event induces a change of the crystal orientation. This result suggests that the crystal growth for the laser ablation is determined mainly around this time scale.

INTRODUCTION

Film deposition by the laser ablation has experienced an exciting development since the successful result of high-Tc film growth by Bellcore group [1]. They demonstrated that the composition of multicomponent complex can be reproduced in the film deposited by the laser ablation. Various techniques have been applied to the film deposition by the laser ablation for enhancing the crystal growth. One of the most interesting techniques is a laser irradiation onto a growing film surface, which was proposed by us, resulting in an improvement of film morphology and superconducting properties [2]. In this experiment, we have been employing only one excimer laser, leading to no delay between the ablation and the irradiation lasers. For optimization of the technique, a variation of the delay time between the two lasers is essential because the film growth by the laser ablation is intermittent. Furthermore research on the effect of the delay time is inevitable for clarifying the mechanism of the deposition by the laser ablation. Here, $Ba_2YCu_3O_x$ superconducting films were prepared by Nd:YAG laser ablation, equipped with a second (ArF excimer) laser for irradiation onto the growing film surface.

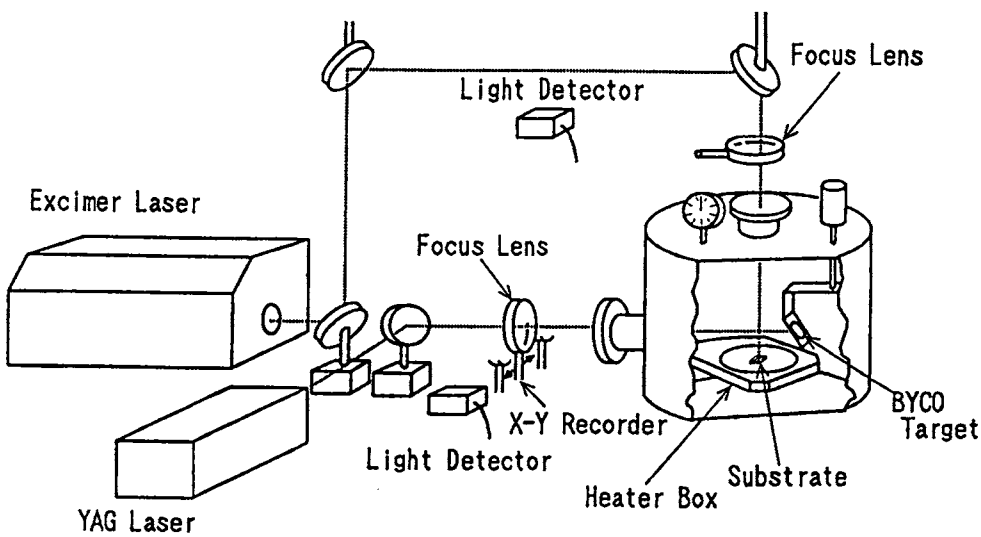

Fig. 1 A schematic diagram of our deposition system

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EXPERIMENTAL

A schematic diagram of our deposition system is shown in Fig.1. Nd:YAG laser was employed for the ablation, and ArF excimer laser for the irradiation. Each laser light was detected by a Si photodiode equipped with a digital storage oscilloscope for determination of the delay time t_d by which the irradiation was delayed after the ablation. Almost all substrates used were unannealed (100) MgO single crystals unless otherwise noted. They are cleaned by the conventional procedure for removing the contaminant. Deposited films at a certain substrate temperature were cooled down to the room temperature at a rate of 4 deg/min in oxygen ambient of a 1.3 kPa. Detailed description on the two lasers is summarized in Table I, and the preparation condition is summarized in Table II.

Table I Lasers used for the ablation and the irradiation

	ablation	irradiation
laser	Nd:YAG laser	ArF excimer laser
type	QUANTEL INT YG661-10	SHIBUYA SQL2240
wavelength	532 nm	193 nm
pulse width	7 ns	10 ns
fluence	5 J/(cm^{2}shot)	50 mJ/(cm^{2}shot)
repetition	5 Hz	5 Hz

Table II Preparation condition

substrate	(100) MgO
target	$Ba_2YCu_3O_X$ pellet
substrate temperature T_S	520-620 °C
oxygen pressure $P(O_2)$	27 Pa

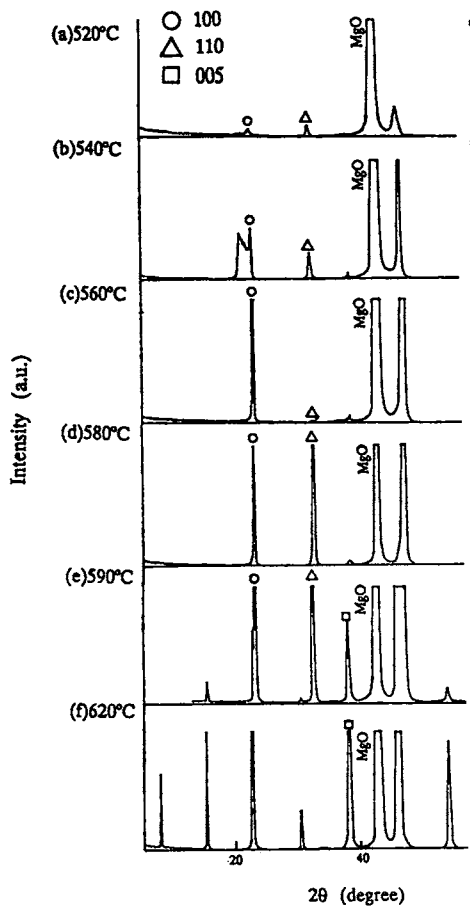


Fig. 2 Substrate temperature dependence of XRD pattern for $Ba_2YCu_3O_X$ films

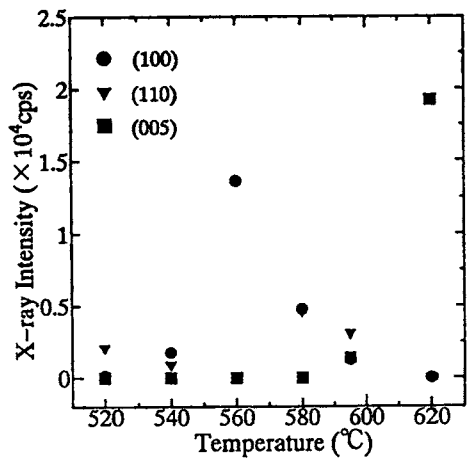


Fig. 3. Substrate temperature dependence of the peak intensities for diffraction peaks from (100), (110) and (005) planes

RESULTS AND DISCUSSION

First of all, we examined the substrate temperature dependence of the films deposited without the second laser irradiation. The target-substrate spacing was set to be 3.6 cm. Figure 2 shows the substrate temperature dependence of X-ray diffraction (XRD) pattern. Almost all the diffraction peaks can be assigned both to the regular MgO and $Ba_2YCu_3O_x$ structures. Diffraction from the substrate is denoted by MgO, and the typical diffractions from $Ba_2YCu_3O_x$ films are denoted by circle (100), triangle (110) and square (005). Remainders were also related diffractions from $Ba_2YCu_3O_x$ structure. Although at low substrate temperatures the diffraction peak originating from (100) dominates the XRD pattern, increasing the substrate temperature enhances the diffraction peak originating from (110). At the highest substrate temperature, there appears the diffraction peak originating from (005). We can also see other diffraction peaks from c-plane in Fig.2(f). Figure 3 shows the substrate temperature dependence of the peak intensities due to diffraction peaks from (100), (110) and (005) plane. This substrate temperature dependence is similar to that reported for sputtered $Ba_2YCu_3O_x$ films [3].

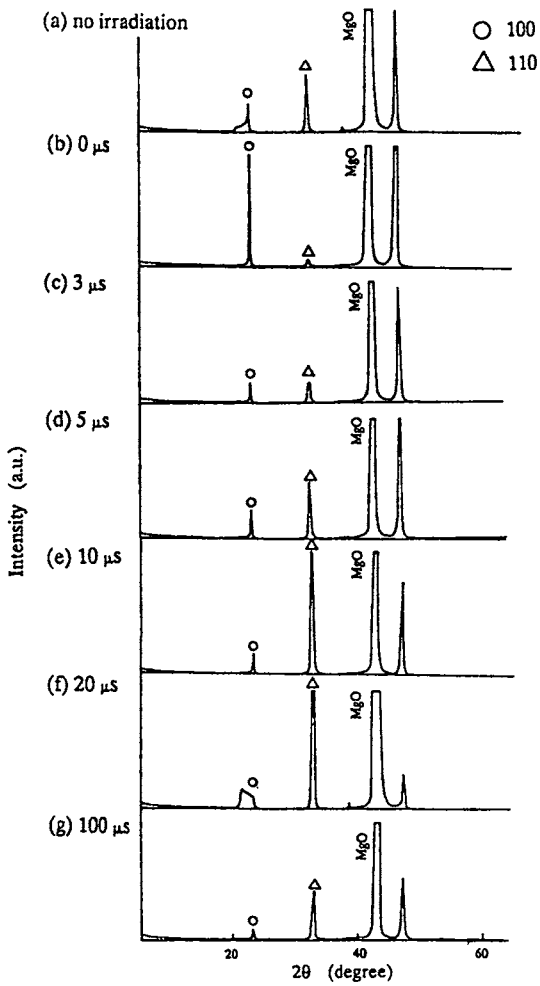


Fig. 4 Delay time dependence of XRD pattern for $Ba_2YCu_3O_x$ films

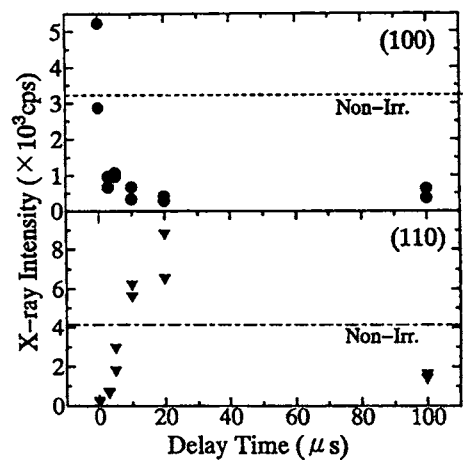
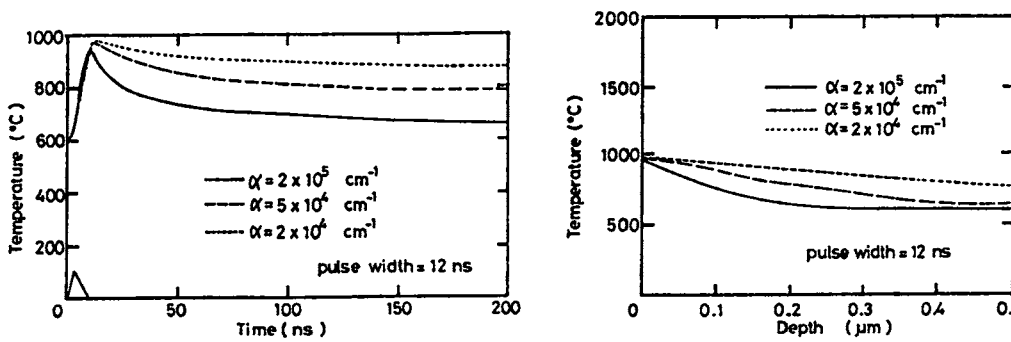


Fig. 5 Delay time dependence of the peak intensities for the diffraction peaks from (100) and (110) planes

Various models for the mechanism of the crystal orientation as a function of the substrate temperature have been proposed. For example, the lattice mismatch between $Ba_2YCu_3O_X$ films and $SrTiO_3$ substrate varies with temperature because lattice constants of both materials varies with temperature due to the thermal expansion and due to oxygen deficiency [4]. Another model is based on a migration mechanism. Present $Ba_2YCu_3O_X$ films have a large anisotropy on the crystal structure. It is rather difficult to grow c-axis oriented film, compared with the other oriented films because c-plane has only one kind of cation atom, leading to a difficulty in migration of those cation atoms for relaxation to a thermal equilibrium site [5]. Former model seems to be inconsistent with this system, because of the large lattice mismatch between MgO and $Ba_2YCu_3O_X$.

Delayed laser irradiation onto a growing film surface was found to have a pronounced effect on the crystal orientation, depending on the delay time t_d. Figure 4 shows XRD patterns of $Ba_2YCu_3O_X$ as a function of t_d. The substrate temperature is 540 °C and the target-substrate spacing is 3.6 cm. Typical diffraction peaks representing respective diffractions are denoted by the circle (100) and the triangle (110). Without the irradiation the (110) diffraction is stronger than the (100) one. The irradiation with the 0 μs enhances the (100) diffraction, and reduces the (110) one on the contrary. However, the irradiation with a finite delay time enhances the (110) diffraction with an increase in the delay time. The irradiation with a delay time of 100 μs reduces the enhancement effect. This result is summarized in Fig.5. This figure shows the delay time dependence of the diffraction peak intensities of the (100) and (110) planes. Dashed and dot-dashed lines represent (100) and (110) peak intensities for films without the irradiation, respectively. This figure clearly shows that an optimum delay time for (110) orientation is several tens μs.

Similar result was observed in our another experiment. In the experiment, the target-substrate spacing is 2.5 cm, and the substrate temperature is 560 °C. It also shows a presence of an optimum delay time around 2 μs for (110) orientation. Thus the optimum delay time has positive correlation with the target-substrate spacing, suggesting that the second laser irradiation may affect the ejected particles arriving at the substrate. The arrival time of the ejected particles can be estimated to be in the order of μs - tens μs for a target-substrate spacing of a few cm based on the data reported by Eryu et al.[6]. They reported that there are four components with various velocities. For example, the arrival time of the second and the third components are estimated to be μs - tens μs for the present target-substrate spacing. Thus this estimation does not contradict the above model. Of course, we cannot neglect a transient heating effect of the film surface and a UV excitation effect of oxygen.



(a) (b)

Fig. 6 Temperature simulation for $Ba_2YCu_3O_X$ film irradiated by a laser;
Transient temperature of the film surface as a function of the time (a) and the depth profile of the temperature when the surface temperature becomes maximum (b)

For evaluating the heating effect, the transient temperature increase of the substrate due to the pulsed laser irradiation was simulated by a computer calculation based on the our previous model [7]. Used characteristic parameters for the calculation are the same ones as the previous model. Here, the substrate temperature is kept at 600 °C . We assumed that the $Ba_2YCu_3O_x$ medium has an infinite thickness, and all the absorbed laser light is converted to heat promptly, and thermal emission from the surface is negligibly small. Figures 6(a) and (b) show the transient temperature of the film surface as a function of the time and the depth profile of the temperature when the surface temperature becomes maximum, respectively. The original point of the time scale in Fig.6(a) represents a starting time of the triangular laser pulse. Based on the absorption coefficient α of crystal $Ba_2YCu_3O_x$ reported by Inam et al. [8], 2×10^5 cm^{-1} appears to be the most reliable one. A laser fluence of 40 $mJ/(cm^2 shot)$ was used for the simulation with $\alpha = 2 \times 10^5$ cm^{-1}. The simulated result in the case of an absorption coefficient of 2×10^5 cm^{-1} shows that maximum temperature increment is around 300 °C, the effect of temperature increase due to the laser irradiation remains even after a few hundreds μs, and the heating effect is mainly limited to a depth of a few hundred nm. These results suggest that the transient heating effect of the film due to the second laser irradiation cannot be negligible.

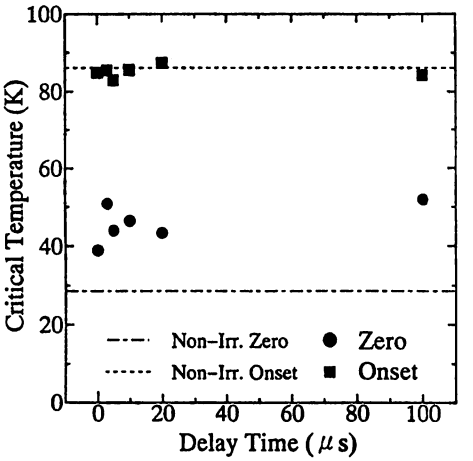


Fig. 7 Delay time dependence of the critical temperature, T_c(onset) and T_c(zero)

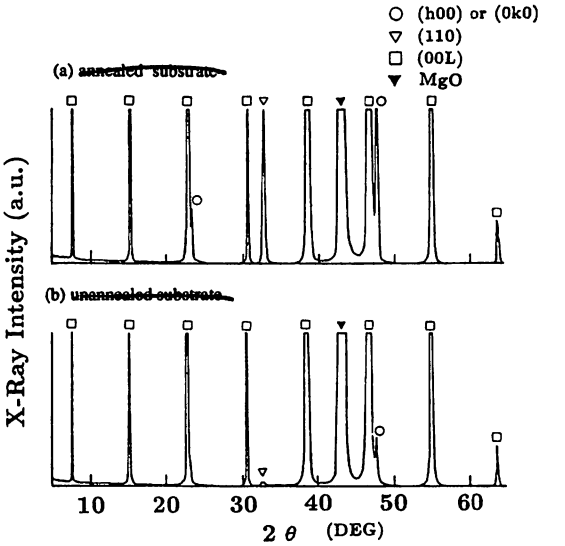


Fig. 8 XRD patterns for $Ba_2YCu_3O_x$ films on the unannealed (a) and annealed (b) MgO substrates

In any events, the experimental result shows that the second laser irradiation within several tens μs around the ablation event induces a change of the crystal orientation. This result suggests that the crystal growth for the laser ablation is determined mainly around this time scale. Furthermore, it was revealed that (110) crystal orientation is enhanced by the second laser irradiation onto the growing film surface with a certain delay time. This result suggests that the laser irradiation enhances the migration of the deposited particles on the film surface, because the change of the orientation from (100) to (110) was observed by increasing the substrate temperature T_S as shown in Figs.2 and 3.

Figure 7 shows the delay time dependence of the critical temperature, T_c(onset) and T_c(zero). Dashed and dot-dashed lines represent the T_c(onset) and T_c(zero) for films without the irradiation, respectively. Although T_c(zero) is really improved by the second laser irradiation, the delay time dependence of it is not so prominent. This finding suggests that the (110) orientation induced by the change of the delay of the second laser does not imply an improvement of grain boundary structures.

Finally we describe the heat treatment effect of MgO substrates on the crystal orientation. All the results described above is reproduced in the experiment if we use the MgO substrate selected from the same lot. However, it becomes difficult to get a reproducible result if we use MgO substrates selected from the various lots. In order to improve the reproducibility, we annealed MgO substrates at 1000 °C for 5 hours in the oxygen ambient of 1 atm. Figure 8 shows the XRD patterns of $Ba_2YCu_3O_X$ films on the unannealed and the annealed MgO substrates. The annealed MgO substrate reduces the orientation other than the c-axis orientation. Not only a c-axis orientation but also an improvement of the reproducibility was realized by this heat treatment of substrates. The detailed result will be published elsewhere.

CONCLUSION

It is shown that the second laser irradiation within several tens μs around the ablation event induces a change of the crystal orientation. This result suggests that the crystal growth for the laser ablation is determined mainly around this time scale.

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