Origin of the Thickness Dependence of Critical Current Densities in YBCO Films Prepared by Pulsed Laser Deposition

K Ohki, K Develos-Bagarinao, H Yamasaki and Y Nakagawa
National Institute for Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568 Japan
kotaro.ohki@aist.go.jp

Abstract. Introducing porosity allows us to increase film thickness of the YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) films on sapphire substrates without microcracking. However, we have observed that porous films possess relatively low values of $J_c$, especially in thick film region (> 500 nm). In order to find out the cause of the degradation of $J_c$, we examined the depth profiles of the magnetic-field angular dependence of critical current density $J_c(t, \theta)$ and other properties for YBCO films ($t \leq 1550$ nm). The YBCO films were deposited by a large-area pulsed laser deposition system on CeO$_2$-buffered r-cut sapphire. Depth profiles were obtained for YBCO films successively milled with an Ar ion beam irradiation. In the as-grown film, the $J_c(\theta)$ exhibits a broad $J_c$ peaks at $H \parallel c$, and smaller one centered at $H \parallel ab$. After the milling, the $J_c(H \parallel ab) / J_c(H \parallel c)$ ratio increases with decreasing film thickness. The YBCO (005) peak of x-ray diffraction shifts towards higher angle with decreasing film thickness. The depth profiles reveal that the oxygen deficit is one of the causes of the decrease of the $J_c$ of the large-area PLD YBCO/CeO$_2$/Al$_2$O$_3$ films with increasing film thickness.

1. Introduction
High temperature superconductor films are expected for use in electric power applications, such as fault current limiters [1] and coated conductors used for various power devices [2]. These electric power applications require a high critical current $I_c$. We have been fabricating large-area YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) films on CeO$_2$-buffered sapphire substrates by a pulsed laser deposition (PLD) method [3], since sapphire has good thermal and mechanical properties with relatively low costs. However, significant reduction of the critical current density $J_c$ with increasing film thickness has often been observed [2-5]. The reduction is generally observed in typical YBCO films, but the YBCO films deposited on sapphire substrates exhibited decay to very low level with increasing film thickness [3]. The $J_c$ decreases more likely to be caused by porosity, which is introduced to suppress micro-cracks [6], but the detailed causes remains unclear.

To clarify the cause of the $J_c$ decrease from point of view of the flux pinning mechanism, several studies have been performed on the large-area PLD (LA-PLD) YBCO thick films with the magnetic-field angular dependence of $J_c$ measurements [7]. The previous study showed that the $J_c$ of the LA-PLD YBCO thin film (<200 nm) exhibits a maximum centered at $H \parallel ab$, but the peaks at $H \parallel c$ became more prominent than the $H \parallel ab$ peaks with increasing film thickness [7]. The angular dependence is peculiar to our films since typical YBCO films show only $J_c$ peaks at $H \parallel ab$. Several
studies have investigated the association between these peaks and the pinning sources. \( H \parallel ab \) peaks are associated with (i) the intrinsic pinning [8,9], (ii) random pinning due to point-like defects [10,11] and (iii) extended planar defects parallel to the \( ab \)-planes [12]. On the other hand, the \( H \parallel c \) peaks are proved to be due to \( c \)-axis correlated pinning centers such as dislocations, planar defects parallel to the \( c \)-axis, and nano-precipitates elongated along the \( c \)-axis [7,13,14]. These results imply that the lack of the pinning centers correlated along the \( ab \) plane is one of the causes of the \( J_c \) decrease. However, the details remain unanswered.

In this study, we obtained the depth profiles. We thinned the YBCO thick films by Ar ion milling and measured the angular dependent \( J_c \) and obtained x-ray diffraction data.

2. Experimental details

2.1. Sample preparation The YBCO films, with thickness up to 1550 nm, were prepared on CeO\(_2\)-buffered (1\( \bar{T} \)02) Al\(_2\)O\(_3\) (\( r \)-cut sapphire) substrates by an LA-PLD system utilizing a KrF excimer laser source (248 nm) operated at an energy density on the target surface of \( \sim 1 \) J/cm\(^2\). The target-to-substrate distance of the LA-PLD system is 14 cm, much longer than standard small-area PLD system. Since the preferential scattering of Ba and Cu occurs, YBCO films deposited from stoichiometric targets are always yttrium-rich. In order to compensate for the deficiency in the films, we used off-stoichiometric targets [6]. The depositions were carried out at an oxygen pressure of 175 mTorr and the substrate temperature of 750 °C. After the deposition, the films were cooled to room temperature in O\(_2\) at 250 Torr.

For comparison, we also prepared YBCO films on (100) SrTiO\(_3\) by a standard PLD system (S-PLD) utilizing an ArF excimer laser source (193 nm). The target-to-substrate distance of the S-PLD system is 3 cm. The YBCO films were deposited at an oxygen pressure of 300 mTorr and the substrate temperature of 760 °C. After the deposition, the film was annealed at 420 °C in O\(_2\) at 700 Torr for 1 h.

The critical temperature \( T_c \) was measured by using a standard four-probe method and magnetic susceptibility measurements. Typical \( T_c \) values of the YBCO/CeO\(_2\)/Al\(_2\)O\(_3\) and the YBCO/SrTiO\(_3\) films were around 88 K and 90 K, respectively. The \( c \)-axis oriented growth was confirmed by x-ray diffraction analysis.

2.2. Depth profiling The depth profiles of \( J_c \) were obtained for YBCO films that were successively milled with an Ar ion beam. The ion beam was produced with an electron cyclotron resonance ion source, and the ion beam irradiation was performed with an acceleration voltage of 500 V. To avoid irradiation damage the samples were cooled by liquid nitrogen during the milling process. The \( J_c \) of the films was measured by an inductive method using third-harmonic voltages [15,16] in liquid N\(_2\) bath (77.3 K). This nondestructive measurement by the inductive method has the advantage that we do not need to make narrow bridges such as necessary in transport measurements. This allows us to make x-ray analysis for each milled films more easily.

2.3. Angular dependence of \( J_c \) The angular dependent \( J_c \) measurement is a very useful tool to investigate pinning sources [7,10,11]. In this study, the angular dependence of \( J_c \) of the YBCO films is also measured by the inductive method [15,16]. A small coil, an outer diameter \( D \) = 2.2 mm, was mounted on a film, and both were fixed on a rotating sample holder. Figure 1 shows the schematic diagram of the coil setup. The angular studies were performed in a liquid N\(_2\) bath (77.3 K) at \( \mu_o H = 0.5 \) T. The angle \( \theta \) is defined as the angle between \( H \) and the normal to the film. This inductive method corresponds to the transport measurement with the field applied perpendicular to the current direction (maximum Lorenz force configuration) [17].

3. Results and discussion
The \( J_c \) of the YBCO films as a function of film thickness up to 1550 nm is shown in Fig. 2. The thickness dependence of as-grown films (filled-circle), which was measured for many different films,
shows a typical degradation of $J_c$ with increasing film thickness. Especially, in thick film region (>500 nm), the $J_c$ exhibits decay to very low values.

In order to investigate inner region of the films, we milled three LA-PLD as-grown films by Ar ion beam irradiation. Two of the three samples are low $J_c$ films and another is relatively high $J_c$ film. Broken lines for samples LA1550, LA610 and LA560 show the thickness dependence of $J_c$ of the thinned films, which were milled from 1550 nm, 610 nm and 560 nm, respectively. After the successive milling, these three films showed only a slight increase of $J_c$ with decreasing film thickness. The $J_c$ of LA1550 increased from 0.24 MA/cm$^2$ to 0.46 MA/cm$^2$, which is much lower than the $J_c$ expected for as-grown thin films.

For comparison, we measured the thickness dependence of $J_c$ in milled ST760 films as shown in open-square. In this sample, the $J_c$ increased with decreasing film thickness and such a recovering phenomenon has been already observed and reported [2]. These results show that the $J_c$ of inner region of the S-PLD film remained high and there is no obvious ion-milling damage. It is suggested that the inner region of LA-PLD films was degraded with further film deposition process possibly due to the growth of large pores.

Figure 3(a), (b) and (c) show the depth profile of the angular dependent $J_c$ of LA1550, LA560 and ST760, respectively. The angular dependent $J_c$ of the LA610 was not shown here because it showed similar tendency to that of LA1550. Basically, the $J_c$ enhanced for the entire angular range with decreasing film thickness, with exception of LA560. The $J_c(H // ab) / J_c(H // c)$ ratio of the LA-PLD films increases with decreasing film thickness. Further, for the thin film region (< 200 nm), $J_c$ peaks at $H // ab$ become prominent. The changes of the $J_c(H // ab) / J_c(H // c)$ ratio are one of the features of the LA-PLD films. Typical YBCO films show $J_c$ peaks at only $H // ab$ as shown in Fig. 3(c), and the characteristic shape of $J_c(\theta)$ of the ST760 did not change for each thickness, except for the film thinned to 150 nm.

We investigated the lattice constant $c$ of the LA-PLD film by YBCO (005) x-ray diffraction peaks. Figure 4 shows the YBCO (005) peaks of the milled film in Fig. 3. As shown in Figs. 4(a) and (b), the YBCO (005) peaks of LA1550 and LA560 shifted towards higher angle with decreasing film thickness. This indicates that the lattice constant $c$ contracted. This contracting of the lattice constant $c$ means the increasing of the oxygen content of the film [18,19]. It is interesting to note that the results indicate that the oxygen content at the interface region is higher than at the surface region. We have also

---

**Fig. 1** Schematic diagram of the coil setup.

**Fig. 2** Thickness dependence of $J_c$ for YBCO films at 77.3 K, self-field: (a) LA560, (b) LA610, (c) LA1550 and (d) ST760. The solid line and broken line indicate the as-grown films and the etched films, respectively.
observed that the $J_c$ ($H // ab$) peak associated with the random pinning increases with oxygenation of the film, which will be described elsewhere [17].

The lattice constants $c$, which are calculated from the YBCO (005) peaks are 11.65 nm, 11.66 nm and 11.70 nm for as-grown ST760, LA560 and LA-PLD 1550, respectively. According to the report [19], there is almost no oxygen deficit in ST760 and very low-level oxygen deficit in LA560. On the other hand, it is estimated that the oxygen deficit $\delta$ of LA1550 is 0.25. It is to be noted that the YBCO (005) peak of the ST760, which correspond to almost no oxygen deficit, did not change with decreasing film thickness (Fig. 4(c)). These results lead us to the conclusion that the oxygen deficiency of the films is one of the causes of the degradation of $J_c$ with increasing film thickness.

In order to increase $J_c$ of the thick films, we tried to increase oxygen content for the YBCO films deposited using the LA-PLD system with the same condition as LA1550 by typical oxygen annealing. However, we could not increase oxygen content of the YBCO films to the level of ST760. The origin of the oxygen deficiency is under investigation.

4. Summary and conclusions

We have studied the depth profiles of the magnetic field angular dependence of $J_c$ and the lattice constant $c$ of the YBCO/CeO$_2$/Al$_2$O$_3$ films deposited by the large-area pulsed laser deposition system. The depth profiles revealed that the oxygen content at the surface region is lower than at the interface region. The oxygen deficiency is one of the causes of the decrease of $J_c$. 

Fig. 3 Angular dependence of $J_c$ for YBCO film at 77.3 K and $\mu_0 H = 0.5$ T. The solid line indicates the data for as-grown films and the broken lines show the data for the milled films LA1550, LA560 and ST760.

Fig. 4 $2\theta$–$\theta$ XRD scans around YBCO (005) peaks of (a) LA1550, (b) LA560 and (c) ST760.
References

[1] Yamasaki H, Furuse M and Nakagawa Y 2004 Appl. Phys. Lett. 85 4427
[2] Foltyn SR, Jia QX, Arendt PN, Kinder L, Fan Y and Smith JF 1999 Appl. Phys. Lett. 75 3692
[3] Develos-Bagarinao K, Yamasaki H, Nie JC and Nakagawa Y 2005 Supercond. Sci. Technol. 18 667
[4] Foltyn SR, Wang H, Civale L, Jia QX, Arendt PN, Maiorov B, Li Y, Maley MP and MacManus-Driscoll JL 2005 Appl. Phys. Lett. 87 162505
[5] Emergo RLS, Wu JZ, Ayug T and Christen DK 2004 Appl. Phys. Lett. 85 618
[6] Develos-Bagarinao K, Yamasaki Nie J C, Murugesan M, Obara H and Nakagawa Y 2005 IEEE Trans. Appl. Supercond. 15 2962
[7] Develos-Bagarinao K, Yamasaki H, Murugesan M, Mawatari Y, Nakagawa Y and Nie J C 2005 Supercond. Sci. Technol. 18 S266
[8] Tachiki M and Takahashi S 1989 Solid State Commun. 70 291
[9] Tachiki M and Takahashi S 1989 Solid State Commun. 72 1083
[10] Civale L, Maiorov B, Serquis A, O Willis J, Coulter J, Wang H, X Jia Q, Arendt PN, MacManus-Driscoll JL, Maley MP and Foltyn SR 2004 Appl. Phys. Lett. 84 2121
[11] Civale L, Maiorov B, MacManus-Driscoll JL, Wang H, Holesinger TG, Foltyn SR, Serquis A and Arendt PN 2005 IEEE Trans. Appl. Supercon. 15 2808
[12] Grigis Ch, Schamm S and Dorignac D 1999 J. Mater. Res. 14 2732
[13] Diaz A, Mechin L, Berghuis P and Evetts J E 1998 Phys. Rev. Lett. 80 3855
[14] Yamada H, Yamasaki H, Develos-Bagarinao K, Nakagawa Y, Mawatari Y, Nie J C, Obara H and Kosaka S 2004 Supercond. Sci. Technol. 17 58
[15] Claassen JH, Reeves ME, J Soulen R and Jr 1991 Rev. Sci. Instrum. 62 996
[16] Yamasaki H, Mawatari Y and Nakagawa Y 2003 Appl. Phys. Lett. 82 3275
[17] Ohki K, Yamasaki H, Develos-Bagarinao K and Nakagawa Y 2007 unpublished
[18] Gallagher P K, O’Bryan H M, Sunshine S A and Murphy D W 1987 Mat. Res. Bull. 22 995
[19] Ueda Y and Kosuge K 1988 Physica C 156 281