Review paper

Flux-pinning mechanisms in high current density superconducting YBa₂Cu₃O₇ filmsHirofumi Yamasaki, Kotaro Ohki¹⁾, Hiroshi Yamada²⁾, Katherine Develos-Bagarinao, Yoshihiko Nakagawa and Yasunori Mawatari

National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan

Fax: 81-29-861-5726, e-mail: h.yamasaki@aist.go.jp

1) Present address: Sumitomo Electric Industries, Ltd., Osaka, Japan

2) Present address: Oshima National College of Maritime Technology, Yamaguchi, Japan

Recently, long coated conductors, in which superconducting YBa₂Cu₃O₇ (YBCO) thin films are deposited on oriented metallic tapes, have been developed, and high critical current densities J_c are required for their application to various power devices. In this review we present our recent investigations on the flux pinning mechanisms in YBCO thin films that determines J_c .

Key words: superconducting thin film, YBa₂Cu₃O₇, critical current density, flux pinning

1. INTRODUCTION

To utilize high-temperature superconducting oxides that show superconductivity above the liquid nitrogen temperature (77 K) for power devices, such as power transmission cables and high-field magnets, long coated conductors, in which superconducting YBa₂Cu₃O₇ (YBCO) thin films are deposited on oriented metallic tapes, have been developed. To achieve large current carrying property, high critical current densities J_c are required, and it is very important to clarify the flux-pinning mechanisms that determine J_c . In this review we present our recent investigations on the flux pinning mechanisms in YBCO thin films.

2. CRITICAL CURRENT DENSITY AND FLUX PINNING

When an electrical current flows through a superconductor, no voltage (resistance) is generated if the current is smaller than a specific value, a critical current, I_c . However, a finite resistance sets in when the flowing current becomes larger than I_c . It is because quantized flux lines, whose core part is in a normal state, enter the superconductor and are moved by the Lorentz force that is the product of the applied magnetic field and the current (Fig. 1). The current flows also in the normal core in the flux-flow state. To increase the critical current density J_c , which is the critical current per unit area of the cross section of the superconductor, we must “pin” the flux lines by introducing appropriate crystal defects, where flux lines stay stably (Fig. 1).

So far many studies have been made on the flux-pinning mechanisms of epitaxial (RE)Ba₂Cu₃O₇ thin films (RE = Y, Nd, Sm, Gd, Dy, Ho, ---; abbreviated as (RE)BCO), in which high J_c values have been observed [1–9]. In early days Hylton and Beasley theoretically pointed out that high J_c observed in YBCO thin films is due to high density of point defects [1]. There were several experimental studies reporting that J_c of YBCO films increased with the density of nanometer-sized Y₂O₃ precipitates and that they are effective pinning centers [2, 3]. As extended defects

correlated along the c -axis of YBCO, screw dislocations formed with the crystal growth of YBCO and twin boundaries that exist in all the superconducting (orthorhombic) YBCO films were sometimes claimed to be primary pinning centers [4–6], but such claims were not supported because there was not good correlation between J_c and defect density. In contrast, Dias et al. clearly showed that edge dislocations in low-angle grain boundaries work as strong c -axis-correlated pinning centers [7]. We also confirmed by magnetic-field angle dependent $J_c(H, \theta)$ measurements and transmission-electron-microscope (TEM) observations that planar defects and dislocations along the c -axis are the effective correlated pinning centers in YBCO films prepared by large-area pulsed laser deposition (PLD) [8, 9]. In recent years development of (RE)BCO coated conductors has been very active. To improve their flux-pinning properties many studies have been performed to introduce artificial pinning centers, such as nano-rods and nano-particles by adding impurity atoms like Zr [10–12]. Nonetheless, until very recently the origin of naturally-formed “natural pins”, which become the primary pinning centers in most (RE)BCO films, were not clear, and we were not able to answer a simple question, “Why high J_c is observed in epitaxial (RE)BCO thin films?”.

Fig. 1 Quantized flux lines and flux pinning.

3. EXPERIMENTAL

3.1 Thin films used for the $J_c(H, \theta)$ measurements

We prepared epitaxial YBCO thin films with the c -axis normal to the film surface on $\text{LaAlO}_3(100)$ single crystals and CeO_2 -buffered sapphire ($\alpha\text{-Al}_2\text{O}_3$) substrates by PLD methods [8, 9, 13]. We used two different systems, one of which is a standard system with target-to-substrate distance $D = 60\text{--}70$ mm and another is a large-area deposition system with $D = 112\text{--}142$ mm. We also investigated the flux-pinning properties in commercial large-area YBCO/ CeO_2 /sapphire thin films, which were prepared by a thermal co-evaporation method [14] and used for fault-current limiter applications [15, 16]. The parameters of the YBCO films described in this article are summarized in Table I.

Table I Parameters of YBCO films for which angular dependent J_c was measured. Substrate, thickness d , critical temperature T_c , self-field J_c at 77 K and the FWHM of (005) plane diffraction peaks in the rocking curves $\Delta\omega$. The T_c values were measured as onset temperatures of diamagnetic ac susceptibility.

Film #	Substrate	d (nm)	T_c (K)	J_c (MA/cm ²)	$\Delta\omega$ (deg)
1	$\text{LaAlO}_3(100)$	150	87.6	2.0	0.49
2	$\text{CeO}_2/\text{Al}_2\text{O}_3$	200	87.4	1.1	1.21
3	$\text{CeO}_2/\text{Al}_2\text{O}_3$	170	91*	3.5	0.49
4	$\text{CeO}_2/\text{Al}_2\text{O}_3$	400	~ 87.5	1.65	0.39
5	$\text{CeO}_2/\text{Al}_2\text{O}_3$	525	86.1	1.2	0.42
6	$\text{CeO}_2/\text{Al}_2\text{O}_3$	420	88.9	3.0	0.41
7	$\text{CeO}_2/\text{Al}_2\text{O}_3$	480	88.3	2.6	0.35
8	$\text{LaAlO}_3(100)$	500	90.6	5.0	0.33
9	$\text{CeO}_2/\text{Al}_2\text{O}_3$	300	86.6	2.8	0.61

*Zero-resistance temperature.

3.2 Methods for evaluating $J_c(H, \theta)$ and microstructures

We performed magnetic-field angle dependent J_c measurements and TEM observations on many YBCO films, and typical examples are described in this article. The $J_c(H, \theta)$ data for several PLD films for Figs. 3, 5 and 10 were obtained by a standard four-probe transport method for $20\text{ }\mu\text{m} \times 2\text{ mm}$ bridges with an electric-field criterion of $1\text{ }\mu\text{V/mm}$, using the Physical Property Measurement System (PPMS, Quantum Design) [8]. The $J_c(H, \theta)$ data for large-area PLD films for Fig. 8 and a co-evaporated film for Fig. 12 were obtained using a nondestructive ac inductive method with a criterion of $E_c = 1\text{ }\mu\text{V/cm}$, in which the samples were immersed in liquid nitrogen (77.3 K) [17, 18]. The angle θ is defined as an angle between H and the normal to the film. During the J_c measurements, the applied magnetic field was perpendicular to the superconducting current [18]. The film microstructure was examined using a transmission electron microscope (TEM, model JEM-2000EX, operated at 200 kV).

4. c -AXIS-CORRELATED PINNING CENTERS IN YBCO FILMS DEPOSITED BY LARGE-AREA PLD

We have developed YBCO films with a large-area PLD system that can deposit 5 inches diameter area, in which laser beam scanning approach is used (Fig. 2) [19–21]. We use large target-to-substrate distances of 11–14 cm to avoid blocking of the laser beam with the substrate heater, and YBCO films deposited from stoichiometric targets were always yttrium-rich due to the preferential scattering of Ba and Cu [19]. The obtained Y-rich films possessed high density of pores, and films thicker than $1\text{ }\mu\text{m}$ can be grown without microcracks on CeO_2 -buffered sapphire substrates, on which YBCO films with few defects thicker than $\sim 400\text{ nm}$ result in microcracks due to a tensile strain originating from the difference of thermal expansion coefficients between sapphire and YBCO [19]. Although such porosity was considered beneficial to increase film thickness without microcracking, the obtained films had rather low J_c values. We investigated film preparation conditions and demonstrated that an effective control of porosity and composition in YBCO films with using non-stoichiometric targets can enable the attainment of relatively high J_c even for thick films [21].

Figure 3 exhibits angular dependent $J_c(H, \theta)$ data for YBCO thin films with relatively low J_c , prepared in an early stage of our investigation to deposit YBCO films by large-area PLD [19, 20]. Film #2 shows $J_c(\theta)$ peaks around $H \parallel ab$ with the shape of a mountain, which are generally observed in many YBCO films. In contrast, film #1 show relatively sharp $J_c(\theta)$ peaks around $H \parallel c$ in addition to the mountain-like $J_c(\theta)$ peaks around $H \parallel ab$. To identify what kind of defects have caused such $J_c(\theta)$ peaks at $H \parallel c$, we investigated the microstructure of film #1. First, we used an etch pit method as used by Dam et al. [5], and observed high density of irregularly-curved etch grooves due to planar defects parallel to the c -axis [8]. We then performed TEM observations, and it was found that such planar defects along the c -axis cross twin boundaries with an angle of 45° , as shown in a planview TEM image of Fig. 4. These planar defects are most probably stacking faults parallel to the (100) plane or (010) plane, as observed in MOCVD-YBCO thin films [22]. It was concluded that relatively sharp $J_c(\theta)$ peaks at $H \parallel c$ (Fig. 3) are caused by such planar defects parallel to the c -axis [8].

Fig. 2 Schematic diagram of a large-area PLD system in which laser beam is rastered for a large target area.

Fig. 3 $J_c(H, \theta)$ in (a) a YBCO/LaAlO₃ film (#1) and (b) a YBCO/CeO₂/Al₂O₃ film (#2), both prepared by large-area PLD.

Fig. 4 TEM planview image for film #1.

We have developed large-area PLD YBCO films on CeO₂-buffered sapphire substrates for the application to microwave devices and fault-current limiters [15, 16]. We succeeded to improve their J_c properties by controlling the composition using non-stoichiometric targets [21] and by annealing CeO₂ buffer layer at ~1000°C to obtain smooth surfaces [23, 24]. Nonetheless, J_c decreased with film thickness, as shown in Fig. 7 in the next page. This thickness dependence is generally observed in the YBCO film deposition by various preparation methods, such as TFA-MOD [25], photo-assisted MOCVD [26] and PLD [27].

To investigate the origin of the J_c decline with film thickness, we measured $J_c(H, \theta)$ for YBCO films with various thicknesses [9, 28]. Typical examples are shown in Fig. 5. Although high- J_c thin film (#3) showed $J_c(\theta)$ peaks both at $H \parallel ab$ and at $H \parallel c$ with similar heights, rather thick film #4 showed large and sharp $J_c(\theta)$ peaks at $H \parallel c$, and the peak at $H \parallel ab$ was much smaller. In the planview TEM picture of Fig. 6, high density of planar defects and some edge or screw dislocations (dark spots, linear defects $\parallel c$) are observed. We examined the density of planar defects and dislocations by the etch pit

method. The density of dislocations decreased with film thickness whereas that of the planar defects increased considerably. Because $J_c(H \parallel c)$ also decreased with thickness [28], these results suggest that dislocations may be more effective pinning centers than planar defects. Although it was revealed that large and sharp $J_c(\theta)$ peaks at $H \parallel c$ are due to dislocations and planar defects extended along the c -axis of YBCO, the primary cause of the J_c decrease with thickness (Fig. 7) is that the $J_c(\theta)$ peaks around $H \parallel ab$ are much reduced, which is easily understood by comparing Fig. 5a and Fig. 5b. The mountain-like $J_c(\theta)$ peaks around $H \parallel ab$ are mainly due to the random pins (uncorrelated point defects), which will be discussed later.

Fig. 5 $J_c(H, \theta)$ in (a) a thin YBCO/CeO₂/Al₂O₃ film (#3, $d = 170$ nm) and (b) a rather thick film (#4, $d = 400$ nm), prepared by large-area PLD.

Fig. 6 TEM planview image for film #4.

5. MULTILAYER STRUCTURES

To overcome the generally-observed J_c decrease with film thickness we then prepared multilayered structures of YBCO interlayered with thin layers of $\text{DyBa}_2\text{Cu}_3\text{O}_7$ (DyBCO), keeping the thickness of each YBCO layer less than ~ 300 nm [29, 30]. As is shown in Fig. 7, J_c drastically increased for YBCO/DyBCO multilayer films, compared with YBCO monolayer films. The angular-dependent $J_c(\theta)$ of the YBCO/DyBCO multilayer films exhibited a very prominent peaks around $H \parallel ab$ (Fig. 8). It is in clear contrast to the $J_c(\theta)$ of rather thick monolayer film #4 (Fig. 5b). Multilayer approach succeeded to increase J_c of large-area PLD films in “thick-film” regions (≥ 400 nm), which was attributed mainly to the enhanced random pinning [30].

Fig. 7 J_c as a function of film thickness for YBCO monolayer films and multilayer films prepared on CeO_2 -buffered sapphire substrates by large-area PLD.

Fig. 8 $J_c(H, \theta)$ in a monolayer film (#5), a bilayer film (#6) and a trilayer film (#7) at (a) 0.5 T and (b) 2 T.

6. RANDOM PINS

6.1 Large and small random pins

Recently we investigated the field-angle dependent $J_c(\theta)$ in YBCO thin films containing high density of nanometer-sized precipitates, and found that the $J_c(\theta)$ behavior depends on the size of nano-precipitates (random pins) [31]. Planview TEM images such as Fig. 9 showed that film #8 prepared by standard PLD contained high density of nano-precipitates, whose diameters are 6–25 nm. We measured $J_c(H, \theta)$ for film #8, and high, broad $J_c(\theta)$ peaks centered at $H \parallel c$ were observed (Fig. 10). In contrast, in a commercial Y-rich co-evaporated YBCO film containing very small nano-precipitates, whose diameters are smaller than 7 nm (Fig. 11), $J_c(\theta)$ peaks around $H \parallel ab$ with the shape of Mount Fuji were observed (Fig. 12).

The diameter of quantized flux lines is $\sim 2\xi$ (Fig. 1), where ξ is the Ginzburg-Landau coherence length. Because $2\xi_{ab}$ of YBCO films #8 and #9 are estimated as 8–10 nm at 77 K [32], the different $J_c(\theta)$ behavior in Figs. 10 and 12 originates from whether nano-precipitates are smaller or larger than 2ξ . In this sense we call very small nano-precipitates ($< 2\xi$) small random pins, and relatively large ones ($> 2\xi$) large random pins. Theoretical background of this matter is described later in the next subsection.

Fig. 9 Relatively large nano-precipitates found in a YBCO/ LaAlO_3 film (#8) prepared by standard PLD.

Fig. 10 $J_c(H, \theta)$ in a standard PLD-YBCO/ LaAlO_3 film (#8), $\times$ denotes a theoretical curve due to Eq. (5).

Fig. 11 Very small nano-precipitates found in an Y-rich co-evaporated YBCO film (#9).

Fig. 12 $J_c(H, \theta)$ in a co-evaporated YBCO/CeO₂/Al₂O₃ film (#9), $\times$ denotes a theoretical curve due to Eq. (4).

6.2. Scaling of $J_c(H, \theta)$ curves and theoretical analyses based on core-pinning interactions

In the previous subsection it was experimentally shown that nano-precipitates result in Mt-Fuji like $J_c(\theta)$ peaks around $H \parallel ab$ (Fig. 12) when the size is very small ($< 2\xi$), or broad $J_c(\theta)$ peaks centered at $H \parallel c$ (Fig. 10) when the size is larger ($> 2\xi$). We then performed a scaling analysis using an angle-dependent parameter

$$\varepsilon(\theta) = (\cos^2\theta + \sin^2\theta/\gamma^2)^{1/2} \quad (1)$$

to reveal that the angular dependence due to small random pins (Fig. 12) originates from the physical anisotropy of YBCO, $\gamma = 5-7$ [31]. When the $J_c(H, \theta)$ data of Fig. 12 were plotted as a function of the scaled magnetic field $H_{scal} = \varepsilon(\theta)H$ with $\gamma = 7$, many $J_c(\varepsilon H)$ curves collapsed into a single scaled curve (Fig. 13). This clear scaling behavior indicates that the flux-pinning of this film is dominated by small random pins, uncorrelated point defects smaller than 2ξ , and the theoretical background is described below.

Fig. 13 Scaling analysis of $J_c(H, \theta)$ curves of Fig. 12.

We considered elementary pinning interactions in YBCO films based on the core pinning interaction, which originates from a local change in the superconducting condensation energy at the pinning site [31]. When a flux line passes through a pinning center (pin), its energy is changed. If the pin is a non-superconducting particle of size (diameter) $a < 2\xi$ and of volume V , the flux line can lower its energy by $(\mu_0/2)H_c^2V$ if it contains one pin, where H_c is a thermodynamic critical field. The pinning force needed to separate the flux line from the pin can be calculated by dividing the energy gain by ξ , which is the range over which the core changes [33]. An elementary pinning force per pinning center can therefore be expressed as

$$f_p = \mu_0 H_c^2 V / 2\xi. \quad (2)$$

If the size of the pinning center a is larger than 2ξ , the energy gain is $(\mu_0/2)H_c^2\pi\xi^2a$ when the flux line positions itself along a diameter of the pin (Fig. 14). If the pinning center is spherical, the length of intersecting flux line continuously decreases from a to zero when it moves away from the pin. In this case, the appropriate interaction distance is a , and the elementary pinning force is expressed as [33]

$$f_p = (\mu_0/2)H_c^2\pi\xi^2. \quad (3)$$

To obtain the flux pinning force density $F_p = J_c B$ per unit volume, we need the summation of the elementary pinning forces. In the case of *strong flux pinning centers*, the direct summation of elementary pinning forces can quantitatively explain experimental results [34]. If the direct summation approach is used, then Eq. (2) for small pinning centers ($a < 2\xi$) yields the expression, $F_p = f_p N_p = \mu_0 H_c^2 V N_p / 2\xi$, where N_p is the density of pinning centers. The critical current density is calculated as $J_c = F_p / B = H_c^2 V N_p / 2\xi H$. As the angular dependent coherence length along the Lorentz-force direction is expressed as $\xi(\theta) = \xi_{ab}(\cos^2\theta + \sin^2\theta/\gamma^2)^{1/2} = \xi_{ab}\varepsilon(\theta)$ (Fig. 14) [35], we obtain the following expression:

$$J_c = H_c^2 V N_p / 2 \xi_{ab} \varepsilon(\theta) H \propto 1/\varepsilon(\theta) H. \quad (4)$$

The magnetic-field dependence of $J_c(\varepsilon H)$ of Fig. 13 can be reproduced well by Eq. (4) for $\mu_0 \varepsilon H \geq 0.2$ T, and the crosshatched curve ($\times$) in Fig. 12, angular dependence of $J_c \propto 1/\varepsilon(\theta)$, reproduced the experimental data very well.

Similar analysis can be done for pinning centers larger than 2ξ [Eq. (3)]. Assuming a direct summation model, then we obtain

$$J_c = H_c^2 \pi \xi_{ab} \xi(\theta) N_p / 2H = H_c^2 \pi \xi_{ab}^2 N_p \varepsilon(\theta) / 2H \propto \varepsilon(\theta) / H, \quad (5)$$

which is proportional to $\varepsilon(\theta)$, in contrast to the $1/\varepsilon(\theta)$ dependence of Eq. (4). The plot of $\varepsilon(\theta) J_c(0^\circ)$ (crosshatched curve in Fig. 10) semi-quantitatively reproduced the angular dependence of J_c at $\mu_0 H = 0.5$ T. These simple theoretical models based on the direct summation of core pinning interactions and angular dependent coherence length $\xi(\theta)$ have successfully explained the scaling for $J_c(\varepsilon H)$ and the different $J_c(\theta)$ behavior due to the size of nano-precipitates.

Fig. 14 (left) Interaction of a flux line and a large random pin. (right) Lorentz force exerted on a slant flux line.

7. OTHER YBCO FILMS

We have demonstrated that nanometer-sized precipitates are the “natural pins”, which are the origin of high J_c of the YBCO thin films studied. We next considered that such very small nano-precipitates (≤ 7 nm), as observed in Fig. 11, are the “natural pins” in many other high- J_c YBCO films, because the $J_c(\theta)$ peaks around $H \parallel ab$ similar to the Mt.-Fuji-like $J_c(\theta)$ peaks in Fig. 12 are generally observed in YBCO films. Then the first author (HY) measured $J_c(H, \theta)$ in YBCO thin films prepared by a fluorine-free (FF) metal-organic deposition (MOD) method, which were prepared by another group in our institute, AIST [32]. He observed $J_c(\theta)$ curves with shapes relatively resembling to the Mt.-Fuji-like $J_c(\theta)$, and parts of the $J_c(\varepsilon H)$ curves corresponding to the angle range of $\sim 6^\circ \leq |\theta - 90^\circ| \leq \sim 20^\circ$ coincided to form a single scaled curve. Very small nano-precipitates were searched, but only low density of large ones (> 20 nm) were observed. Instead, considerably high density of stacking faults ($\parallel ab$) were observed. He concluded that the dislocations surrounding stacking faults are the “natural pins” that dominate the flux pinning in the FF-MOD thin films

[32]. The dislocation ($\parallel ab$), a strong linear pin whose diameter is smaller than 2ξ , acts as a ab -plane-correlated pin when it is perpendicular to the current direction, and acts as a small random pin in other cases, because the crossing point between the linear pin and the flux line becomes a pin.

He and co-workers also measured $J_c(H, \theta)$ for commercial coated conductors, and strongly suggested that dislocations surrounding stacking faults ($\parallel ab$) play an important role in the flux pinning in commercial MOCVD-YBCO tapes [36]. Such investigations on the “natural pins” are still in progress.

8. SUMMARY

We made extensive investigations on flux-pinning mechanisms in YBCO films prepared by large-area PLD by angular dependent $J_c(H, \theta)$ measurements and TEM observations. We demonstrated that planar defects (most probably stacking faults parallel to the YBCO (100) plane or (010) plane) and dislocations along the c -axis are effective correlated pinning centers. We found that J_c decreased with film thickness, and that this decrease is considerably overcome by a multilayering approach.

We then investigated the flux-pinning properties in YBCO thin films containing high density of nanometer-sized precipitates, which are examples for the “natural pins”. The $J_c(\theta)$ behavior depends on whether the nano-precipitates are larger or smaller than 2ξ , the diameter of flux lines. It has been experimentally and theoretically demonstrated that the Mt.-Fuji-like $J_c(\theta)$ peaks around $H \parallel ab$ and the clear scaling ($J_c(H, \theta) = J_c(\varepsilon H)$) are caused by point defects whose diameters are smaller than 2ξ (small random pins). We also explained why broad $J_c(\theta)$ peaks centered at $H \parallel c$ are observed for YBCO films containing large random pins ($> 2\xi$).

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