



J_c enhancement and flux pinning in $Y_{1-x}Gd_xBCO$ and (Gd,Eu) codoped $Y_{0.9-y}Eu_yGd_{0.1}BCO$ thin films by TFA-MOD

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ABSTRACT

$Y_{1-x}Gd_xBa_2Cu_3O_{7-\delta}$ ($0 \leq x \leq 0.5$) thin films were prepared on $LaAlO_3$ (001) substrates by trifluoroacetate metal organic deposition (TFA-MOD). It was observed that the critical current density under self-field of $Y_{0.9}Gd_{0.1}Ba_2Cu_3O_{7-\delta}$ thin film was about two times higher than that of YBCO thin film within the range of 5–85 K. Then, codoped $Y_{0.9-y}Eu_yGd_{0.1}BCO$ thin films ($0 \leq y \leq 0.2$) were fabricated by TFA-MOD. It was observed that the critical current density under magnetic fields was enhanced largely due to codoping. The flux pinning type was not changed with Gd doping and codoping, which could be attributed to the normal surface pins.

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1. Introduction

$YBa_2Cu_3O_{7-\delta}$ (YBCO)-based coated conductors (CCs) have been explored intensively due to potential applications under high magnetic fields [1,2]. Till now several routes have been verified successfully to enhance the critical current density J_c , among which substitution of Y ions by rare earth element is a common way [3–6]. It has been approved that substitution of Y ions by rare earth element can enhance J_c effectively including YBCO ceramics and YBCO bulks fabricated by melt-textured growth method [5–8], YBCO thin films prepared by pulsed laser deposition and metal organic chemical vapor deposition [9–14].

Trifluoroacetate metal organic deposition (TFA-MOD) as a popular method to prepare coated conductors has been explored widely due to its low cost as well as easy processing of long tapes [15–20]. Currently, it is reported that Gd doping can influence the J_c of YBCO thin films prepared by TFA-MOD. However, the variety of J_c is not quite the same due to different processing parameters [21]. Kaneko et al. found that the J_c of $Y_{1-x}Gd_xBa_2Cu_3O_{7-\delta}$ thin films is improved slightly as compared to that of YBCO thin films [22]. As for $Y_{0.8}Gd_{0.2}BCO$ thin films, it is observed that the J_c under high magnetic fields is higher than that of YBCO thin films, however the J_c under low magnetic fields is lower for $Y_{0.8}Gd_{0.2}BCO$ thin films [23]. Moreover, substitution of Y ions by two different types of rare earth elements (codoping) has been proved as an effective route to enhance J_c in bulks such as (Nd,Eu,Gd) BCO [24,25]. However,

there have no reports about codoping results in YBCO thin films by TFA-MOD methods.

In this paper, $Y_{1-x}Gd_xBCO$ thin films were prepared firstly by TFA-MOD, and it was observed that the J_c under self-field was about two times for the $Y_{0.9}Gd_{0.1}BCO$ thin film as compared to that of YBCO thin film within the temperature range of 5–85 K. Then, in order to further enhance performances under magnetic fields, YBCO thin films codoped by Eu and Gd were fabricated, and it was found that the codoping can obviously enhance the J_c under magnetic fields.

2. Experimental

$Y_{1-x}Gd_xBa_2Cu_3O_{7-\delta}$ (YGBCO, $0 \leq x \leq 0.5$) and $Y_{0.9-y}Eu_yGd_{0.1}Ba_2Cu_3O_{7-\delta}$ (YEGBCO, $0 \leq y \leq 0.2$) thin films were prepared by TFA-MOD. Y (Gd, Eu), Ba, and Cu acetates were dissolved into trifluoroacetic acid and deionized water, and the solutions were stirred for 10 h at room temperature. Then, the solutions were refined by distillation process to yield blue glassy residues, which were then dissolved into methanol. The solutions were refined for three times in order to remove water, TFA and acetic acid impurities. Finally, methanol was added into the solutions to adjust the solution concentration to 2.5 M in cations.

All thin films were prepared by spin coating method onto $LaAlO_3$ (001) (LAO) single crystal substrates using rotation speed of 4500 rpm for 60 s. The deposited thin films were pyrolyzed in flowing humid oxygen (the water vapor percentage of the gas is 4.2%). Then, the pyrolyzed thin films were crystallized at 800 °C in flowing humid 500 ppm oxygen/nitrogen atmosphere. Finally, the thin films were annealed at 450 °C in dry oxygen to absorb oxygen.

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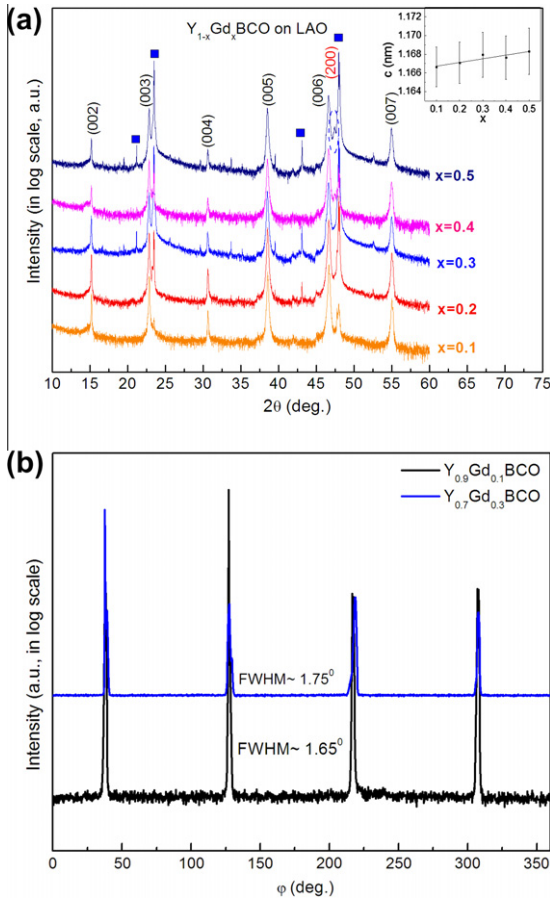


Fig. 1. (a) XRD θ - 2θ patterns of all YGBCO thin films and the inset shows the c -axis lattice constant with Gd variation. (b) Phi-scanning patterns of $Y_{0.9}Gd_{0.1}BCO$ and $Y_{0.7}Gd_{0.3}BCO$ thin films.

The microstructures were analyzed by X-ray diffractions to check up the crystallization and orientation, and field-emission scanning electronic microscopy (FE-SEM) to check up the surface morphologies and thickness. The properties were measured by the magnetic property measurement system with a SQUID magnetometer. The J_c was calculated using the extended Bean's model.

3. Results and discussion

Fig. 1a shows the θ - 2θ patterns of all derived YGBCO thin films. It is seen that all thin films are pure without undesirable phases, and the crystal structure can be indexed as an orthorhombic phase. It is seen that when Gd content is less than 0.3, the derived thin films are c -axis oriented; however, when Gd content is higher than 0.3, a diffraction peak corresponding to the YBCO (200) peak can be observed, which is further confirmed by FE-SEM morphologies. From the diffraction peak of YBCO (006), the c -axis lattice constant calculated by the Bragg formula is elongated with increasing Gd content as shown in the inset of Fig. 1a, which can be attributed to the larger ion radius of Gd^{3+} (1.053 Å) as compared to that of Y^{3+} (1.019 Å).

Fig. 2 shows the FE-SEM morphologies of all derived thin films. It is observed that when Gd content is less than 0.3, the density of precipitate nanoparticles with grain size of 80–200 nm is increased with increasing Gd content except for the YBCO thin film. However, when Gd content is further increased, the density of precipitate particles is reduced, which is same as previous reports [23,26,27]. Additionally, some a -axis oriented grains with

needle-shape are observed, which is same as the XRD results. The a -axis oriented grain size is enhanced with increasing Gd, indicating the enhanced nucleation/growth. From the cross-section FE-SEM as shown in Fig. 3f, it is obtained that the thin film thickness is about 250 nm.

For YBCO, when the growth temperature is lower or the oxygen partial pressure is higher than the optimal growth conditions, the orientation of the derived thin films will change from pure c -axis orientation to a -axis orientation [28]. The optimal growth condition to grow YBCO is close to the straight line (Hammond-Bormann line) in the $1/T - \log p_{O_2}$ diagram [29]. Compared with the conditions (temperature and pressure) on the line, the optimized condition to prepare GBCO thin film is shifted to lower oxygen pressures and higher temperatures [30], which will result in the appearance of a -axis oriented grains with increasing Gd.

The zero-field cooled (ZFC) magnetization with the applied field $H = 0.001$ T is used to determinate the critical temperature T_c , and the results are shown in Fig. 3a. It is seen that the T_c for all derived thin films are about 90.5 K.

The critical current density J_c is estimated using the extended Bean critical state model [31,32]

$$J_c = \frac{20\Delta M}{a(1 - \frac{a}{3b})} \quad (1)$$

where ΔM is the width of the magnetization hysteresis loop, and a , b ($b > a$) are the width and length of an orthorhombic cross section perpendicular to the applied magnetic field, respectively.

Fig. 3b shows the estimated J_c results at 77 K for all YGBCO thin films with the applied magnetic field parallel to the c -axis of thin films. It is seen that the $Y_{0.5}Gd_{0.5}BCO$ thin film has the lowest J_c , which can be attributed to the appearance of large amounts of a -axis oriented grains [33]. Moreover, the highest current density under self-field $J_c^{self-field}$ is attributed to the thin film of $Y_{0.9}Gd_{0.1}BCO$. However, the J_c decreases rapidly as increasing magnetic fields as compared with other samples. The temperature dependent $J_c^{self-field}$ for the three typical samples YBCO, $Y_{0.9}Gd_{0.1}BCO$ and $Y_{0.7}Gd_{0.3}BCO$ is shown in Fig. 3c. It is seen that the $J_c^{self-field}$ is about two times for the $Y_{0.9}Gd_{0.1}BCO$ thin film as compared with that of the YBCO thin film within the measured temperature range.

Based on the above results, it is observed that the $Y_{0.9}Gd_{0.1}BCO$ thin film has the highest $J_c^{self-field}$ within the measured temperature range. It is reasonable to further investigate the codoping effects. Then, $Y_{0.9-y}Eu_yGd_{0.1}BCO$ ($0 \leq y \leq 0.2$, YEGBCO) thin films are prepared by TFA-MOD method. Fig. 4 shows the XRD patterns for all YEGBCO thin films. It is seen that all thin films are pure and c -axis oriented. Additionally, it is seen that the c -axis lattice constant is elongated with increasing Eu as shown in the inset of Fig. 4, which can be attributed to the larger ionic radius of Eu^{3+} (1.066 Å) as compared to that of Y^{3+} (1.019 Å).

Fig. 5 shows the FE-SEM morphologies for the samples of $y = 0.1$ and $y = 0.15$. The amount of precipitate nanoparticles is decreased as increasing Eu content, and the needle-shape a -axis grains are disappeared. Moreover, for the sample of $y = 0.15$, the grains exhibit a rectangular shape and are well aligned indicating the enhanced epitaxy, which is favorable for the enhancement of $J_c^{self-field}$ [34].

Fig. 6a shows the ZFC magnetization results measured under a 0.001 T applied magnetic field. It is seen that T_c is enhanced slightly from 90.5 K for the $Y_{0.9}Gd_{0.1}BCO$ thin film to 91.5 K for the $Y_{0.75}Eu_{0.15}Gd_{0.1}BCO$ thin film, and the transition sharpness is unchanged obviously with Eu content, which suggests that Eu ions occupy the Y sites [35].

Fig. 6b shows the J_c - B results at 77 K for all derived YEGBCO thin films. It is seen that the $J_c^{self-field}$ is the highest for the $Y_{0.9}Gd_{0.1}BCO$ thin film, and it decreases rapidly with increasing

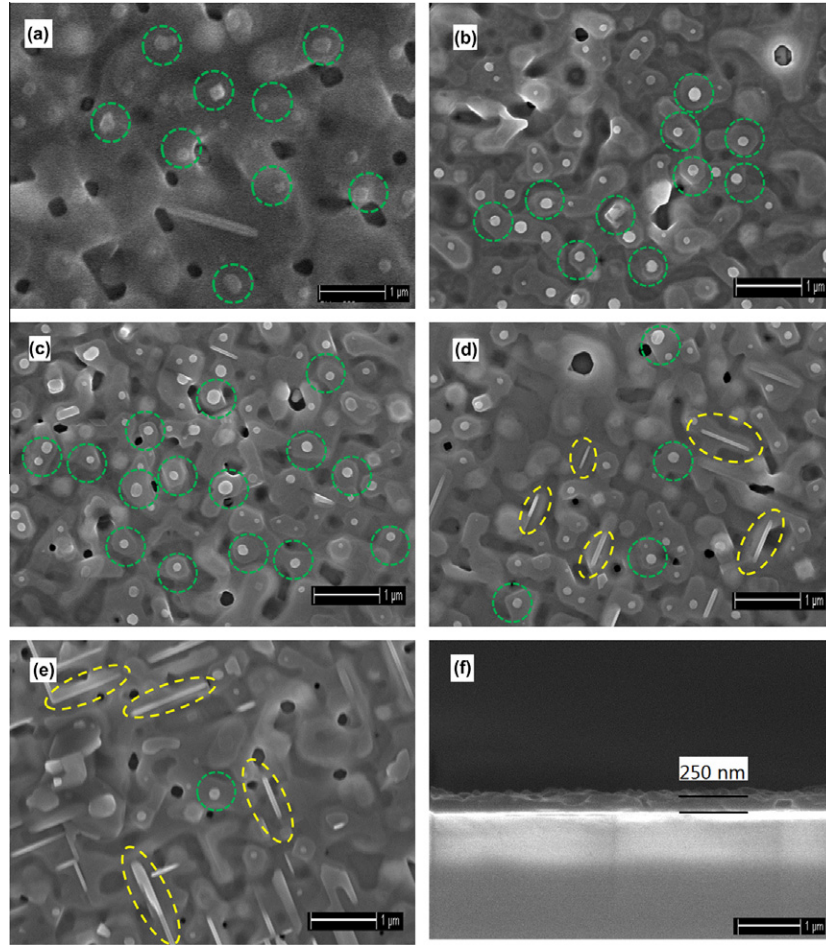


Fig. 2. FE-SEM for YBCO (a), $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ (b), $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ (c), $\text{Y}_{0.6}\text{Gd}_{0.4}\text{BCO}$ (d), $\text{Y}_{0.5}\text{Gd}_{0.5}\text{BCO}$ (e) and cross-section FE-SEM (f) to determine the thickness.

magnetic fields. The J_c for all codoped thin films is higher than that of the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ when the applied magnetic field is higher than 0.02 T. The enhancement of J_c under applied magnetic fields suggests that Eu substitution will introduce effective pinning centers. Fig. 6c shows the detailed J_c results for three typical samples $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$, $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$ and $\text{Y}_{0.8}\text{Eu}_{0.1}\text{Gd}_{0.1}\text{BCO}$ under different applied magnetic fields within 5–85 K.

Fig. 7 shows the double-log J_c – B results at 77 K for three typical Gd-doped thin films and three typical codoped thin films. It is seen that the J_c – B curves can be divided into three regimes. In low fields, a plateau regime is observed, in which J_c is nearly independent of the applied field and can be considered as a strong individual pinning regime; in intermediate fields, a power law regime is suitable and J_c decreases with a power law $J_c \propto B^{-\alpha}$, which is considered as a weak collective pinning regime; finally, J_c drops sharply at high fields due to vortex creep.

In a theoretical B – T phase diagram, in the plateau regime one characteristic field is termed as the accommodation field B_{acc} , marking the crossover from a strong individual pinning to a weak collective pinning. It is also called as the interaction field, below which the vortex–vortex interactions are negligible and J_c is expected to be field independent [36]. The B_{acc} is estimated using the criterion of $J_c(B_{acc})/J_c(0) = 0.90$. It is seen that the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film has the lowest B_{acc} (~ 0.006 T) although its $J_c^{self-field}$ is the highest. Furthermore, for the codoped thin films, the B_{acc} is enhanced to the value of 0.02 T, which is higher than that of the YBCO thin film (0.01 T).

By fitting, the values of α in the power law regimes can be obtained, and the α is 0.77, 0.78 and 0.74 for the YBCO, $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$

and $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$, respectively. These values are higher than that of YBCO thin films prepared by pulsed laser deposition ($\alpha \sim 0.5$ – 0.6), which can be attributed to the lower density of c -axis-correlated dislocations in TFA-MOD derived thin films [16]. It has been shown that the dislocations in the TFA-MOD derived thin films do not connect from the bottom to the top due to lateral grain growth (grain boundary meandering). Moreover, for the codoped thin films, the value of α is decreased to about 0.65.

In double-log J_c – B results, another characteristic field is termed as B_{cr} , which can be determined by the onset of J_c deviation from the power law. At 77 K, the B_{cr} value is the lowest for the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film (~ 0.3 T), while the value is ~ 0.6 T for both the YBCO and $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ thin films. Furthermore, for the codoped thin films the B_{cr} is increased to 0.8 T.

It is observed that at 77 K the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film has the highest value of $J_c^{self-field}$. However, the B_{acc} and B_{cr} are the lowest and the α is almost same as that of the YBCO and $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ thin films, which will lead to the rapid decrease of J_c with increasing applied magnetic fields. For the codoped thin films, the relatively higher $J_c^{self-field}$, larger B_{acc} , and smaller α value will lessen the decay rate, which will lead to a higher J_c with increasing applied magnetic fields.

Fig. 8a shows the volume pinning force $F_p = J_c \times B$ at 77 K for several Gd and codoped thin films. It is seen that the F_p is the largest for the $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ thin film in the series of Gd-doped thin films although the $J_c^{self-field}$ is the highest for the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film. For the codoped thin films, the F_p is improved obviously, which suggests that the flux pinning is enhanced by codoping.

Kramer [37] and Dew-Hughes [38] have proposed a scaling model of flux pinning force, $f(h) = A(h)^p(1 - h)^q$, to characterize

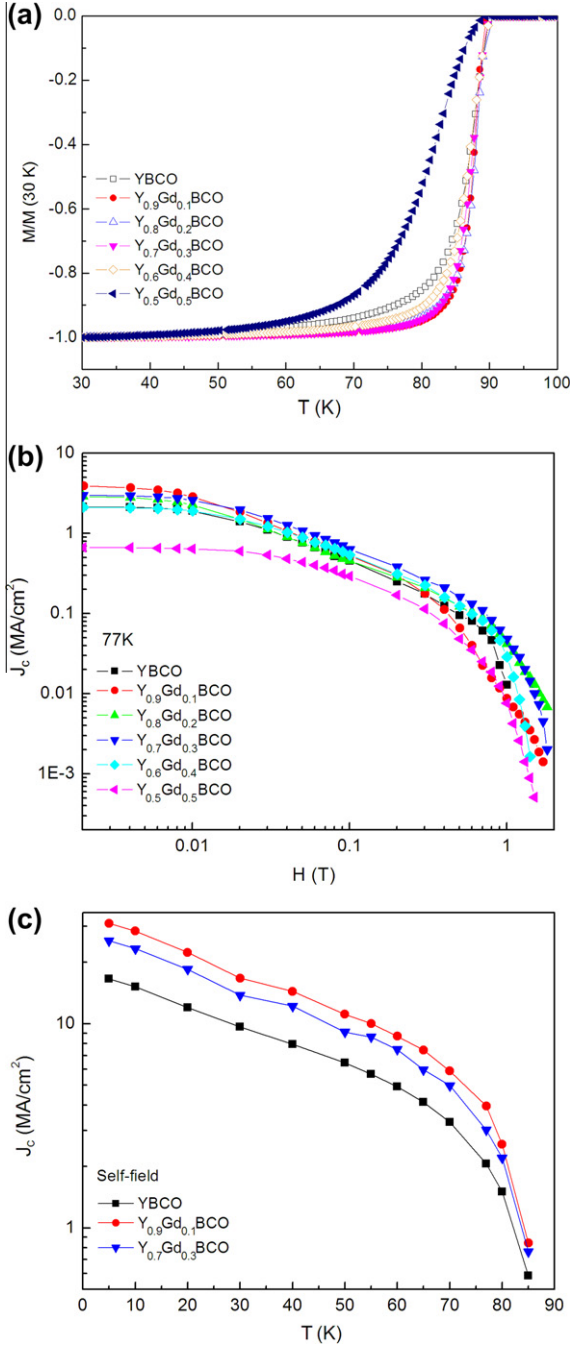


Fig. 3. (a) Normalized M ($M/M(30\text{ K})$) as a function of T for YBCO thin films with applied field of 0.001 T parallel to c -axis. (b) Double-log J_c – B at 77 K results for YBCO thin films. (c) $J_c^{\text{self-field}}$ – T results for three typical samples of YBCO, $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ and $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ thin films.

the pinning mechanism, where $f = F_p/F_{p,\text{max}}$, $h = H/H_{\text{irr}}$, and $F_{p,\text{max}}$ is the maximum pinning force and H_{irr} is the irreversibility field. Fig. 8b shows the normalized volume pinning force at 77 K. The irreversibility field H_{irr} is determined by fitting experimental data using $f(h) = A(h)^p(1-h)^q$. The position of the maximum f is given by $h_0 = p/(p+q)$. As shown in Fig. 8b by fitting, it is obtained that the values of p and q are 0.42 and 1.97, 0.46 and 2.13, 0.50 and 1.89, 0.50 and 1.50 for $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$, $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$, $\text{Y}_{0.8}\text{Eu}_{0.1}\text{Gd}_{0.1}\text{BCO}$ and $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$, respectively. These values are very close to the values of $p = 0.5$ and $q = 2$, which is corresponding to the normal surface pins [39].

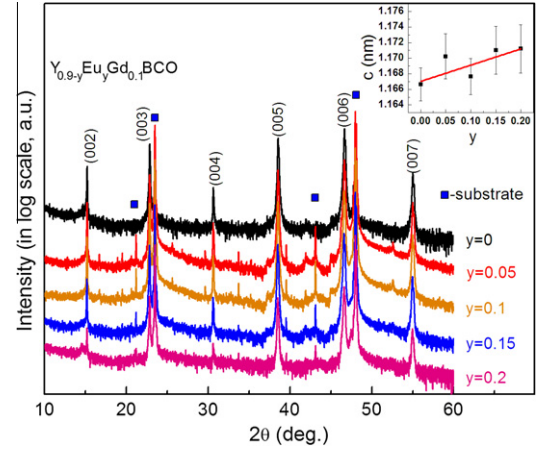


Fig. 4. XRD θ – 2θ patterns of all YEGBCO thin films and the inset shows the c -axis lattice constant with Eu variation.

In type II superconductors, the origination of pinning is originated from the spatial variation of the Ginzburg–Landau (GL) order parameter $|\psi|$; the spatial variation of $|\psi|$ can be associated with the disorders in the T_c and/or with the spatial variations in the charge carrier mean free path l . These two kinds of pinning are called as δT_c and δl pinning, respectively. In both cases J_c can be described as $J_c \propto (1 - t^2)^n$ with $t = T/T_c$. For a single vortex pinning, the characteristic exponent $n = 7/6$ holds for the δT_c pinning or $5/2$ holds for the δl pinning [40]. Fig. 8c shows the t dependent J_c in double-log scale under self-field for three typical samples. It is seen that the obtained n is 1.24, 1.25 and 1.28 for the YBCO, $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ and $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$ thin films within the range of 20–80 K respectively, closing to the value of $7/6$, which indicates that the δT_c -pinning is dominant in all derived thin films. The dominant δT_c -pinning can be attributed to the nonsuperconducting nanoparticles and the strain fields [40]. In general, the strain fields are as a result due to the lattice misfit between the nanoparticles and REBCO, which will induce weak superconducting regions within the matrix [40]. Additionally, nanoscaled lamellar arrays or nanostripes, formed by nanoclusters of rare-earth-rich phases as observed in $\text{Nd}_{0.33}\text{Eu}_{0.38}\text{Gd}_{0.28}\text{Ba}_2\text{Cu}_3\text{O}_y$ and $(\text{Sm}_{0.33}\text{Eu}_{0.33}\text{Gd}_{0.33})\text{Ba}_2\text{Cu}_3\text{O}_y$ bulks [41,42] as well as in YBCO single crystal [43], will lead to the δT_c -pinning.

Under self-field, it is seen that the highest $J_c^{\text{self-field}}$ is attributed to the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film. It has been reported that partial substitution of Y ions by rare earth such as Gd can enhance the $J_c^{\text{self-field}}$ since GBCO has higher superconducting properties as compared with that of YBCO [44], which can explain the higher $J_c^{\text{self-field}}$ for $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ as compared to that of YBCO. On the other hand, as Gd content is further enhanced, the orientation will be degraded as confirmed by ϕ -scanning measurements shown in Fig. 1b, which will lead to the degradation of $J_c^{\text{self-field}}$. As for the codoped thin films, it is seen that the values of $J_c^{\text{self-field}}$ at 77 K are smaller than that of the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film, which can be attributed to the higher superconducting properties for GBCO thin films as compared with that of EBCO. On the other hand, the $J_c^{\text{self-field}}$ at 77 K for the $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$ thin film is higher than that of the $\text{Y}_{0.8}\text{Eu}_{0.1}\text{Gd}_{0.1}\text{BCO}$ thin film, which can be attributed to the excellent epitaxy as shown in Fig. 6.

Upon applied magnetic fields, the J_c is decreased rapidly for the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film due to the larger α value, small B_{acc} and B_{cr} . As estimated from pinning force scaling, it is suggested that the dominant flux pinning is normal surface pins. From the FE-SEM morphologies, it is observed that the amounts of precipitate nanoparticles in the $\text{Y}_{0.7}\text{Gd}_{0.3}\text{BCO}$ thin film are larger than that of the

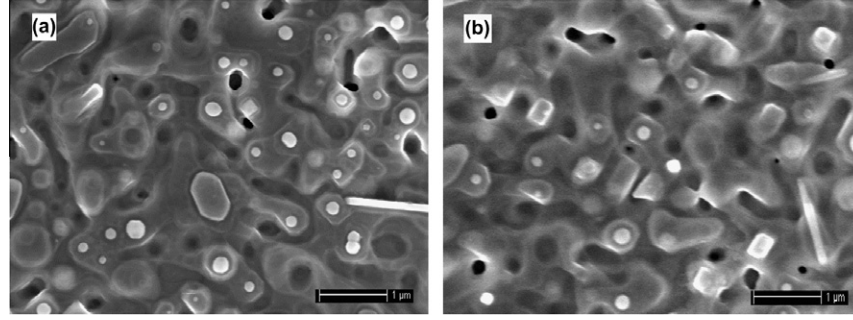


Fig. 5. FE-SEM of $\text{Y}_{0.8}\text{Eu}_{0.1}\text{Gd}_{0.1}\text{BCO}$ (a) and $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$ (b).

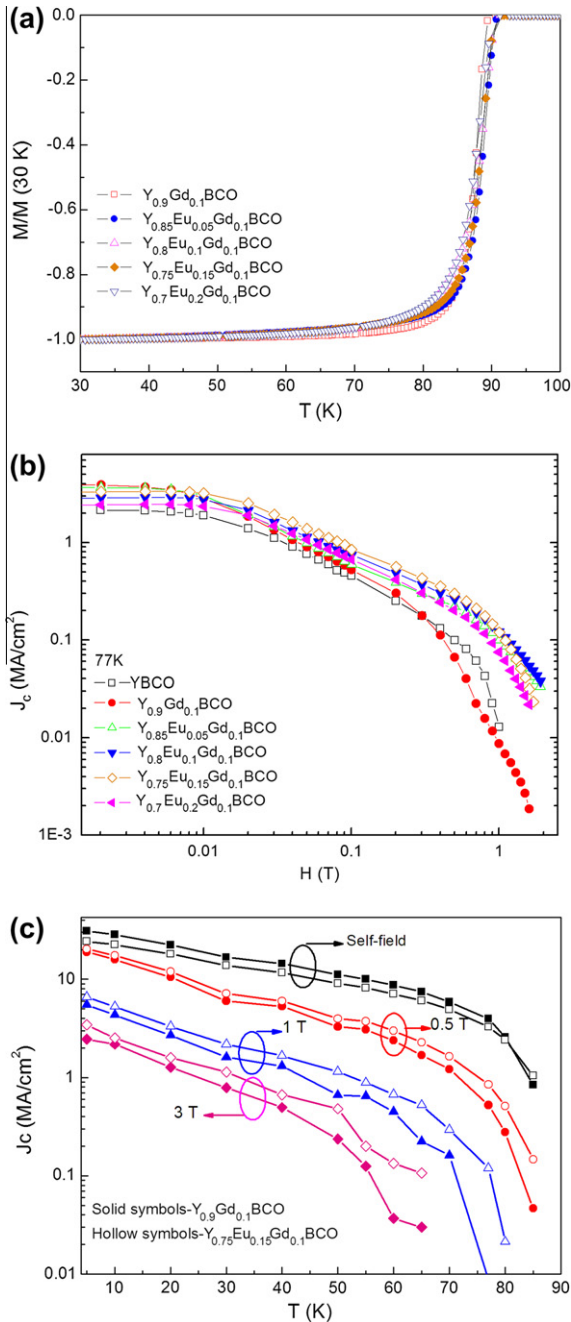


Fig. 6. (a) Normalized $M/M(30\text{ K})$ as a function of T for YEGBCO thin films with applied field of 0.001 T parallel to c -axis. (b) Double-log J_c - B at 77 K results for several typical thin films. (c) J_c - T under different applied magnetic fields for two typical samples of $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ and $\text{Y}_{0.75}\text{Eu}_{0.15}\text{Gd}_{0.1}\text{BCO}$ thin films.

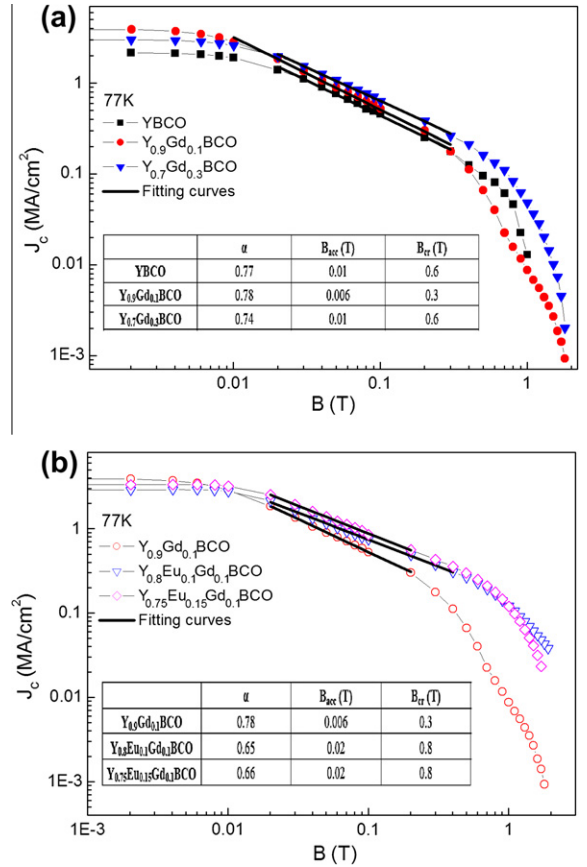


Fig. 7. (a) Double-log J_c - B at 77 K results for three typical Gd-doped thin films and the inset table is the obtained parameters by fitting. (b) Double-log J_c - B at 77 K results for three typical codoped thin films and the inset table is the obtained parameters by fitting.

$\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$ thin film, which will enhance the flux pinning force resulting in the enhanced J_c under applied magnetic fields. However, the amounts of precipitate nanoparticles onto the surfaces of the codoped thin films, in which the J_c is higher than that of the $\text{Y}_{0.9}\text{Gd}_{0.1}\text{BCO}$, are smaller than these of the Gd-doped thin films. That is to say other pins should play a dominant role. Additionally, the similarity in α , B_{acc} and B_{cr} for all codoped thin films suggests that the pins should be originated from same contributions, which maybe originated from the nanoparticles and nanoscaled lamellar arrays within the superconducting matrix. Additionally, the stress field pinning due to ionic radius mismatch in Y sites may also enhance the flux pinning, which can be estimated through the rare earth ion size variance defined as $\sigma^2 = \sum x_i \langle r_i \rangle^2 - \langle RE_{ave} \rangle^2$, where i = rare earth ion, x = RE mole fraction normalized to 1, r is

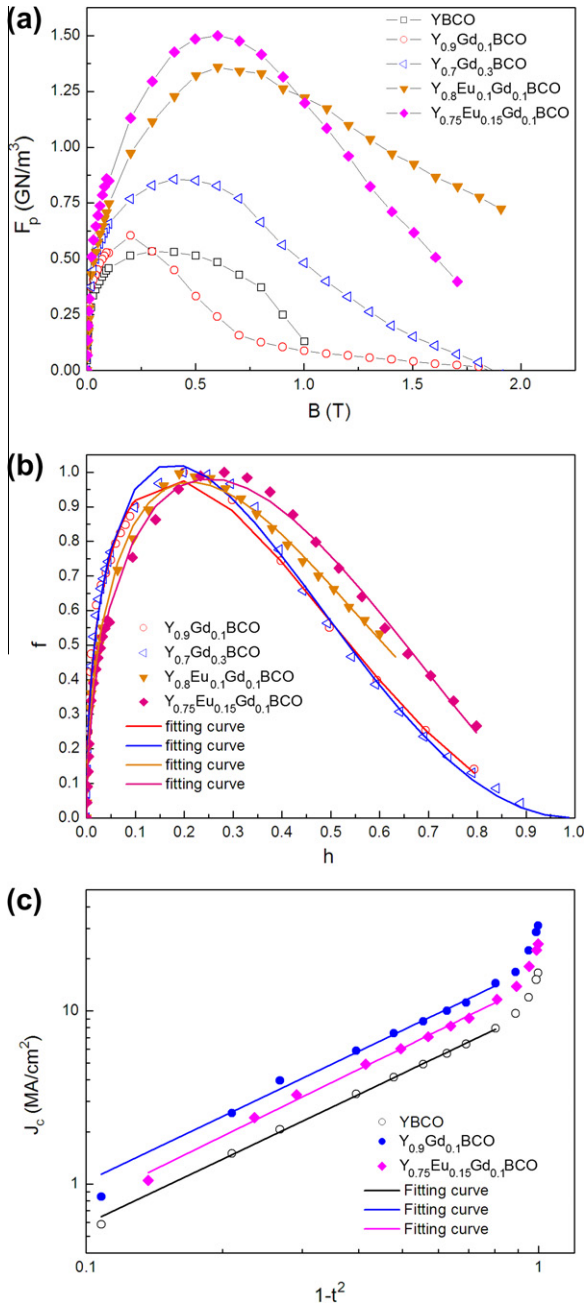


Fig. 8. Volume pinning force (a), Normalized pinning force (b) and Double-log J_c - t (c) for several typical samples.

the ionic RE radii with VIII coordination and +3 valence state, and $RE_{ave} = x_i r_i$ [12]. It is calculated that σ^2 is 0, 1.04×10^{-4} and $2.43 \times 10^{-4} \text{ \AA}^2$ for YBCO, $Y_{0.9}Gd_{0.1}BCO$ and $Y_{0.7}Gd_{0.3}BCO$, respectively. The larger ion size variance will enhance the flux pinning under applied magnetic fields.

4. Conclusions

A series of $Y_{1-x}Gd_xBa_2Cu_3O_{7-\delta}$ ($0 \leq x \leq 0.5$) thin films were prepared on $LaAlO_3$ (001) substrates by trifluoroacetate metal organic deposition. It was observed that the $J_c^{self-field}$ can be enhanced obviously, and the highest $J_c^{self-field}$ was attributed to the $Y_{0.9}Gd_{0.1}Ba_2Cu_3O_{7-\delta}$ thin film with its value about two times higher than that of the YBCO thin film within 5–85 K. Then, codoped $Y_{0.9-y}Eu_yGd_{0.1}BCO$ thin films ($0 \leq y \leq 0.2$) were fabricated and it

was observed that the critical current density under magnetic fields was enhanced largely. The flux pinning type was not changed by doping and codoping and could be attributed to the normal surface pins.

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