In-Field Transport Properties at Grain Boundaries in $BaHfO_3$-doped $SmBa_2Cu_3O_y$ Bicrystal Films at Low Temperatures

Y. Tsuchiya, J. Akita, Y. Ichino, S. Miura, and Y. Yoshida

***Abstract*—High-temperature superconducting REBCO tapes are attractive for their high critical current density J_c in magnetic fields. However, J_c in the REBCO film is suppressed under magnetic fields and by grain boundaries. Our group has reported that artificial pinning centers (APC) enhances the J_c at the grain boundaries in the REBCO films at the liquid nitrogen temperature of 77 K. In this study, we extended our previous work to a lower temperature since the usage of the REBCO tapes at a lower temperature and a high magnetic field is a recent development trend. Transport characteristics at grain boundaries in the REBCO films with and without APC were investigated at temperatures down to 4.2 K and at magnetic fields up to 9 T. The results show that the introduction of the APCs is effective to enhance the J_c at the grain boundaries at low temperatures and at the magnetic fields especially around the matching field where the density of the APC is equal to that of vortices.**

***Index Terms*—Critical current, Grain boundaries, Coated conductors, Cuprates**

I. Introduction

The high-temperature superconducting $REBa_2Cu_3O_y$ (*RE* = rare earth elements, REBCO) coated conductors is one of the promising conductors for application with superconducting magnets such as scientific high field magnets [1] or dipole magnets for accelerators [2]. For the magnet application, the critical current density J_c under a magnetic field needs to be improved. The keys to improve in-field J_c in the REBCO tapes are as follows: enhanced flux pinning using artificial pinning centers (APC) [3] and biaxial orientation using a buffer layer [4].

A number of researches on the APC in the REBCO films have been reported so far [5]-[10]. Non-superconducting perovskite oxide $BaMO_3$ (*M*: metal elements, BMO) is chemically stable in the REBCO matrix and grows like a rod by self-organized growth [5]. Since the BMO rod has good matching with the vortex core, the in-field J_c in the REBCO film is largely enhanced [6]. The metal elements such as Zr, Sn, and Hf have been reported as the *M* in the BMOs forming nanorods [6]-[8]. The $BaHfO_3$ (BHO) nanorod is one the most effective APC to enhance the in-field J_c. Our group has reported the high flux pinning force density F_p in BHO-doped $SmBa_2Cu_3O_y$ (SmBCO) films as 1.5 TN/m^3 at 4.2 K and 21 T [9] and 32.5 GN/m^3 at 77 K and 5 T [10].

A grain boundary (GB) is another key factor to determine the J_c in the REBCO films [11]-[18]. The REBCO tape are biaxially oriented because the J_c at the GBs (J_c^{GB}) exponentially decreases compared with in-grain J_c when a misorientation angle increases [11]-[14]. The J_c^{GB} starts to decrease at a critical misorientation angle (θ_c) [15]. On the other hand, several kinds of research revealed that the J_c^{GB} is equal to the in-grain J_c at high magnetic fields at 77 K [16]-[18]. To enhance in-field J_c^{GB}, it is is necessary to improve the flux pinning of an Abrikosov-Josephson (A-J) vortex with an anisotropic core [19].

It is practically interesting whether the introduction of the APCs improves the in-field J_c^{GB} in REBCO films. So far, the J_c^{GB} characteristics in the REBCO films with the APCs has been reported only at 77 K [20], [21]. However, little is known about the in-field J_c^{GB} at low temperatures. In this study, the J_c^{GB} characteristics in REBCO films with and without the nanorod APCs are investigated at low temperature down to 4.2 K and high fields up to 9 T.

II. Experimental Method

Undoped and BHO-doped SmBCO films were prepared by using the pulse laser deposition. The fabrication method is similar to our earlier report [21]. The films were fabricated with a KrF excimer laser (λ = 248 nm), at 10 Hz repetition rate, with an energy density of 1.0 J/cm^2, at an O_2 partial pressure of 400 mTorr, and at a substrate temperature of 870 °C. All the films had a thickness of 120 nm. BHO was doped into the films using the alternating-targets technique [22]. The BHO-doped films have content of BHO of 4.5vol.% which was determined by the inductively coupled plasma mass spectroscopy. The films were fabricated on (001) $(LaAlO_3)_{0.3}$-$(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$

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Y. Tsuchiya, J. Akita, Y. Ichino, Y. Yoshida is with the Department of Electrical Engineering, Nagoya University, Aichi, 464-8603, Japan (e-mail: tsuchiya@nuee.nagoya-u.ac.jp)

S. Miura is with the Institute of Superconductor Systems, Kyushu University, Fukuoka 819-0395, Japan

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TABLE I
SUPERCONDUCTING PROPERTIES OF THE FILMS.

Undoped/ BHO-doped	Misorientation Angle [°]	T_c [K]	J_c (77 K, self-field) [A/cm²]
Undoped	0 (single crystal)	92.3	5.46
	5	91.8	2.45
	10	91.7	0.20
	15	92.2	0.08
BHO-doped	0 (single crystal)	90.6	2.69
	5	90.7	2.01
	10	90.5	0.29
	15	90.0	0.14

(LSAT) single crystal substrates and [001]-tilted bicrystal LSAT substrates with misorientation angles θ of 5°, 10°, and 15°. The films were processed into micro-bridges including the GBs with a width of 50 μm and a length of 1 mm by using the laser etching technique.

The current-voltage characteristics of the films were measured under a magnetic field B of 0 to 9 T and at a temperature T of 4.2 to 77 K by using PPMS (Quantum Design) and the four-probe method. During the measurement, the magnetic field was applied parallel to the c-axis of SmBCO which includes the GB plane. Applied current flowed perpendicular to the GBs. The non-ohmic linear differential region in which a voltage is not proportional but linearly different was observed for the films with $\theta > 0°$. J_c was determined with an electric field criterion of 1 and 10 μV/cm for the single crystal and the bicrystalline films, respectively. T_c was determined from the temperature-dependent resistivity with a criterion of 0.1 μΩ·cm.

III. Results and Discussion

A. Specification of films

Out-of-plane X-ray diffraction (XRD) showed diffraction peaks of (00l) (l = 1, 2, 3, 4, 5, 6, 7, 8, and 9) SmBCO for all the fabricated films and an additional peak of (200) BHO only for the BHO-doped films. In-plane XRD for the bicrystalline films showed 2 strong diffraction peaks of (102) SmBCO whose difference was equal to the misorientation angle of the bicrystalline substrates. The result indicates the existence of the GB with a misorientation angle of θ. Similar results were obtained for the BHO-doped films.

Tab. 1 shows the T_c and the J_c at 77 K and a self-field in the undoped and the BHO-doped SmBCO films. The T_cs in the BHO-doped films are slightly degraded by the BHO doping. On the other hand, the J_c drastically decreases by one order of magnitude or more for $\theta > 5°$.

B. In-field flux pinning properties

Fig. 1 shows field dependences of the J_cs in the undoped and the BHO-doped films with θ = 0-15° at T = 4.2, 77 K, and at B = 0-9 T. When θ = 0°, it represents the film grown on a single crystal substrate. The J_c decreases with increasing θ at all fields for both the undoped and the BHO-doped films as reported in earlier studies [21]. The J_c drastically drops at θ between 5° and 10°, which indicates the transition from a low-angle GB to a high-angle GB. The in-field J_c in the BHO-doped films were larger than that in the undoped films, suggesting that the J_c^{GB} was improved by the BHO doping into the SmBCO films. The BHO-doped films with θ = 0° and 5° showed plateaus of J_c at B = 0.5-2 T at 77 K. Let us compare the results of the transport characteristics with our previous results [21]. The field dependence of the J_c measured at 77 K is similar to that reported in our

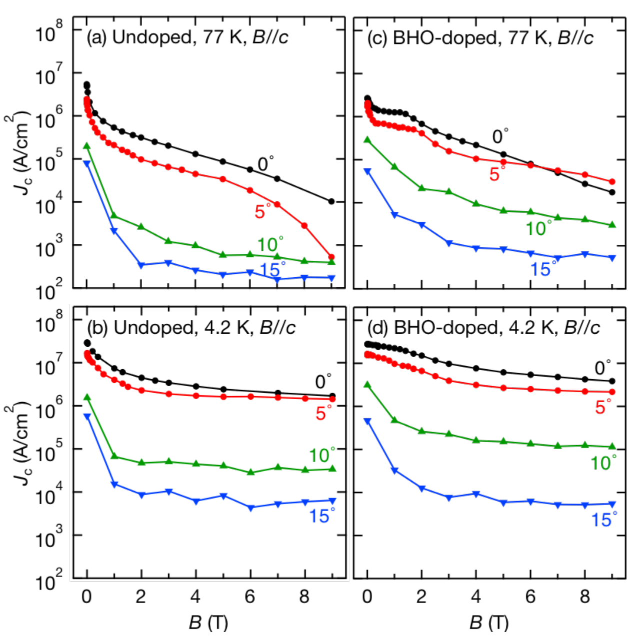

Fig. 1. Field dependence of J_c at grain boundary in the undoped and the BHO-doped SmBCO films at 77 K and 4.2 K.

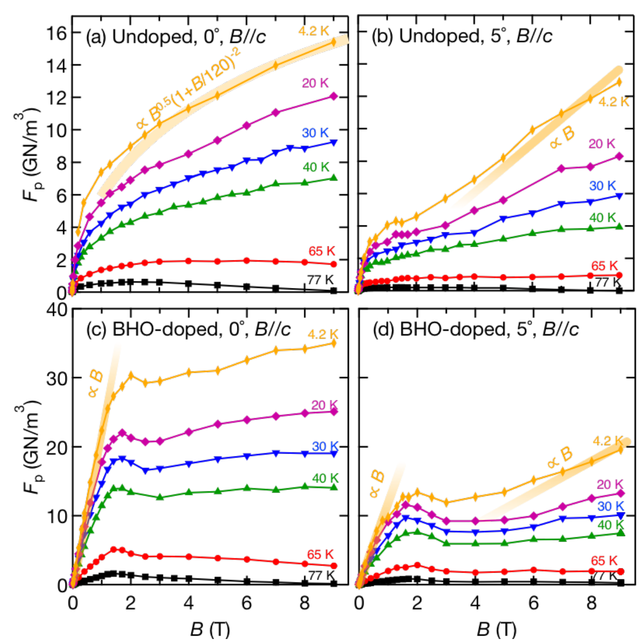

Fig. 2. Field dependence of F_p at grain boundary in (a, b) the undoped and (c, d) the BHO-doped SmBCO films with θ = (a, c) 0°, (b, d) 5° at T = 4.2-77 K.

previous study in terms of the plateau of the in-field J_c^{GB}. The plateaus of the J_c can be ascribed to the existence of *c*-axis oriented APCs in the films. In fact, according to the previous study, the transmission electron micrography clarified that the BHO-doped film had the BHO nanorod APCs which grew parallel to the *c*-axis of SmBCO in the grains and also at the GBs. Therefore, the similar field dependence of the J_c indicates that the BHO nanorods APCs are most likely grown in the films for this study. Turning now to the J_c properties at a low temperature, the J_c at 4.2 K is enhanced by the addition of BHO for $\theta = 0°$, 5°, and 15° as shown in Fig. 1(b) and (d). For example, the J_c^{GB} for $\theta = 5°$ was enhanced 1.5 times by the doping of the BHO APC. This result revealed that the introduction of the APC is effective to improve the J_c^{GB} even at a temperature less than 77 K. As a result different from the high temperature, the plateaus of the J_cs became less obvious. In the followings, we focus on the J_c properties for $\theta = 0°$ and 5° including the temperature dependence to discuss the origin of the enhanced flux pinning properties by the BHO doping.

Fig. 2 shows field dependences of the macroscopic flux pinning force densities F_ps ($= J_c \times B$) in the undoped and the BHO-doped SmBCO films with $\theta = 0°$, 5° at various temperatures ranging from 4.2 to 77 K. As shown in Fig. 2(a), the F_p in the undoped film with $\theta = 0°$ has upward convex curves for all the temperatures. To clarify the origin of the flux pinning for the in-grain J_c in the undoped film, the field dependence of F_p is calculated based on the collective vortex pinning model and is shown as the orange curve in Fig. 2(a) [23]. Here, the typical field dependence of F_p as $F_p(B) \propto B^{0.5}(1 - B/120)^2$ was assumed [9]. The field dependence of the F_p at a low temperature such as 4.2 K and at $B > 4$ T follows the calculated line, which indicates that the in-grain J_c in the undoped SmBCO film with $\theta = 0°$ is explained with the collective flux pinning by the random flux pinning centers.

Next, as shown in Fig. 2(b), linear field dependences of the F_p at $B > 2$ T are observed in the undoped SmBCO film with $\theta = 5°$, which is obviously different from the upward convex field dependence for the in-grain flux pinning. For comparison, an orange straight line proportional to the magnetic field is drawn in Fig.2(b). The linear field dependence of the F_p is explained by the correlated flux pinning model [23]. Diaz has reported that the A-J vortex at the GB is pinned by the dislocation in the bicrystalline YBCO film [24]. Therefore, the dislocation at the GB is a possible origin for the correlated flux pinning. As an evidence to support this discussion, the linearity of F_p to the magnetic field was more obvious at lower temperatures, which indicates that the flux pinning by the dislocation at the GBs is more effective at lower temperatures where the coherence length of the SmBCO is close to the size of the dislocation core (~1 nm) [24]. Let us estimate the field range in which the flux pinning by the dislocation core at the GB is effective. The maximum field is estimated to be 30 T for the dislocations at the GBs in the REBCO films at $\theta = 5°$ based on the dislocation distance of $d_{\rm dis} = a/\sin(\theta/2)$, where a is the lattice constant of the REBCO matrix [13]. In order to prove this estimate, a further J_c measurement at a much higher field is desired.

In the BHO-doped SmBCO film with $\theta = 0°$ as shown in Fig. 2(c), peaks of F_p at $B \sim 2$ T and saturations of F_p at $B > 3$ T were observed at all the temperatures. The field for the F_p peaks corresponds to that of the end of the plateaus in the field dependence of the J_c as shown in Fig. 1(c). At magnetic fields below the peak fields, the F_p is proportional to the magnetic field. The peak of the F_p has been observed in the REBCO films with the straight and continuous nanorod APCs [15], [26]. This feature shows that the density of the BHO nanorods corresponds to the peak magnetic field of ~2 T.

Finally, in the BHO-doped SmBCO film with $\theta = 5°$ as shown in Fig. 2(d), linear magnetic field dependences of the F_p with different slopes are observed in two magnetic field ranges. The peaks of F_p appear at $B \sim 2$ T similar to the BHO-doped SmBCO films with $\theta = 0°$. In addition, linear field dependences also appear at $B > 4$ T at $T < 20$ K similar to the undoped SmBCO film with $\theta = 5°$. Therefore, the field dependence of the F_p in the BHO-doped film with $\theta = 5°$ is explained by the combination of the flux pinning due to the dislocation at the GBs and by the BHO nanorods. The results indicate that the A-J vortex at the GB is pinned more strongly with the BHO doping. Two possibilities are considered for the enhanced flux pinning of the A-J vortices. One is that the A-J vortex is directly pinned by the BHO nanorod grown at GB. The other is the A-J vortex is indirectly pinned by the interaction with the pinned vortices in the SmBCO grains with the BHO nanorods. Further study is needed to clarify which origin is valid.

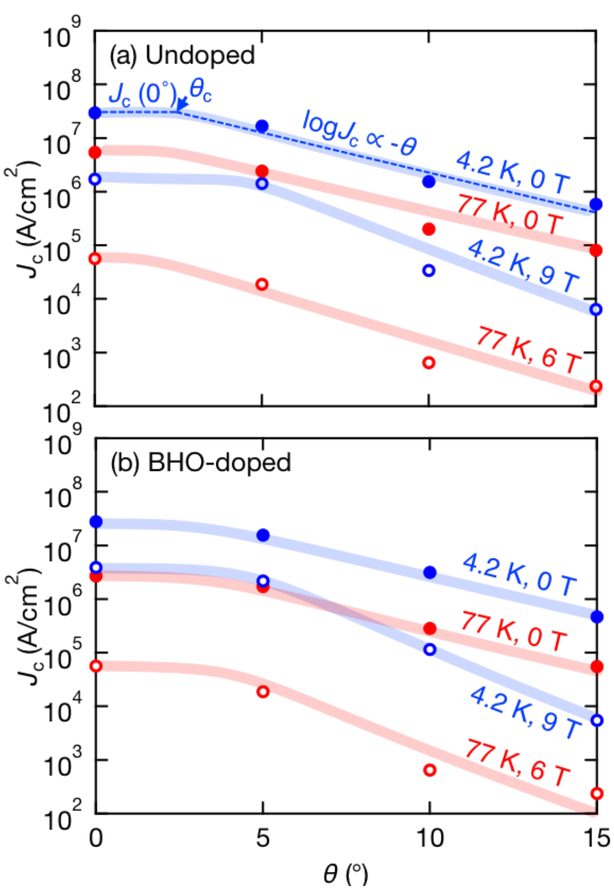

Fig. 3. Misorientation angle θ dependence of J_c^{GB} at conditions of (T, B) = (4.2 K, 0 T), (4.2 K, 9 T), (77 K, 0 T), and (77 K, 6 T) in (a) the undoped and (b) the BHO-doped SmBCO films.

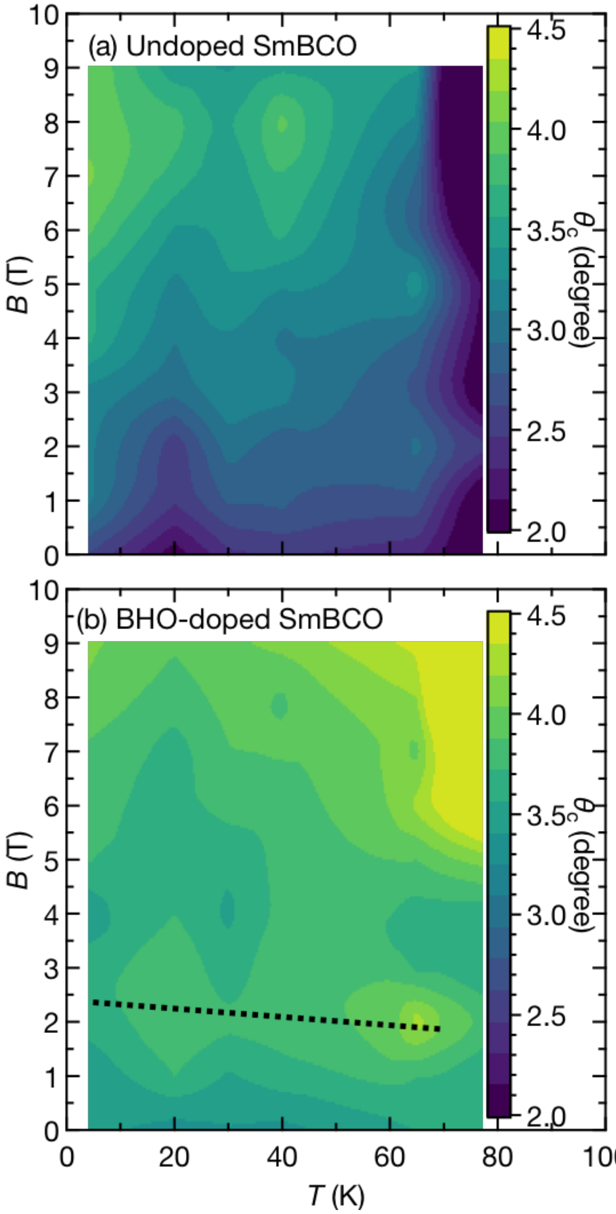

Fig. 4. Mappings of the critical misorientation angle θ_c for magnetic fields and temperatures in (a) undoped and (b) BHO-doped SmBCO films.

C. Critical misorientation angle

Fig. 3 shows θ dependences of the J_c^{GB} in the undoped and the BHO-doped SmBCO films at conditions of $(T, B) = (4.2$ K, 0 T), (4.2 K, 9 T), (77 K, 0 T), and (77 K, 6 T). For all the conditions, the J_c^{GB} monotonically decreases with increasing θ. According to the earlier study [15], the J_c^{GB} starts to decrease from a certain θ which is called as the critical misorientation angle θ_c. In this study, we defined the θ_c as a crossover angle between the constant J_c^{GB} at low-angle GBs and the exponential decrease of the J_c^{GB} at high-angle GBs. The θ_c is an important parameter to describe the allowance of the misorientation, and the larger θ_c is preferable for the application. The θ_c is estimated as follows. Draw a line with a constant J_c from $\theta = 0°$ as shown in Fig. 3(a). Then, a decay of J_c^{GB} for $\theta > 0°$ is fitted with an exponential function. Finally, the θ_c is estimated as a crossing θ for both the lines. B and T dependences of the θ_c is discussed in the following paragraphs.Fig. 4 shows the mapping of the θ_c as a function of B and T for the undoped and the BHO-doped films. The mappings are colored in the same color scale. Overall, the θ_c was higher for the BHO-doped films as 3.2-4.4° than that for the undoped films as 2.0-4.0° for a broad range of B and T. A region with the highly enhanced θ_c was observed at $T > 65$ K and $B > 7$ T. We do not go into detail of this region because it is close to the irreversibility line of the SmBCO films. At a low $T < 20$ K and a high $B > 6$ T, the θ_c is ~ 4° regardless of the BHO doping. This common behavior is explained by the flux pinning by the dislocation at the GB because the correlated flux pinning by the dislocation is effective at low temperatures. Finally, in the BHO-doped films, the θ_c shows a ridge at 2-2.5 T for wide T range as shown as a dotted line in Fig. 4(b). This field corresponds to the end field of the plateau of the J_c as shown in Fig. 1(c) and the peak field of the F_p as shown in Fig. 2(c) and 2(d). The pinning efficiency for the correlated pinning center is maximized in a magnetic field called a matching field where the density of vortices is equal to that of the pinning centers [23]. According to the previous study [21], the enhancement of J_c^{GB} is maximum at the matching field for the BHO-doped SmBCO films. Therefore, the ridge of the θ_c in the BHO-doped SmBCO films in this study is due to the enhanced J_c^{GB} at the matching field for the BHO nanorod APCs. Note that the enhancement of the J_c^{GB} by the BHO doping is slight only for $\theta = 15°$. It is possibly because the A-J vortex at the low-angle GBs is pinned by the APCs but the Josephson vortex at the high-angle GB is not. This result suggests that the θ_c can be further improved by optimizing the flux pinning of the A-J vortices at the GBs such as changing the diameter of the nanorod APCs.

IV. Conclusion

In this study, the low-temperature and the in-field superconducting properties at the GBs in the BHO-doped SmBCO bicrystalline films were investigated. As results, the J_cs at the low-angle GB were improved by the BHO doping at a broad ranges of temperatures and magnetic fields. The θ_c in the SmBCO films were improved especially at a field around the matching field. This result suggests that the BHO doping into the SmBCO film is practical for fabrication of the coated conductors in terms of that the BMO APC doping makes J_c^{GB} less susceptible to the degree of orientation of the buffer layer on the substrate.

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