In-Plane Anisotropy of Transport Property in BaTbO₃-doped SmBa₂Cu₃O_y Films

H. Kato, Y. Tsuchiya, Y. Ichino, A. Ichinose, and Y. Yoshida

Abstract-BaTbO3 (BTbO) is one of artificial pinning center materials and forms thick rectangular nanorods with diameters of ~20 nm when it is doped high. We expected that the rectangular nanorods would produce in-plane anisotropy of flux pinning. In this study, we investigated in-plane anisotropy of transport properties in BTbO-doped SmBa₂Cu₃O_y (SmBCO) films. SmBCO thin films with various contents of BTbO were fabricated by using a pulsed laser deposition method. To investigate the in-plane anisotropy, the critical current density J_c was measured at 77 K, 0-9 T, and $\phi = 0^\circ$ and 45°, where ϕ was defined as an in-plane angle from the *a* or *b*-axes of SmBCO. To fix the direction of current in the films, the microbridges were fabricated with different ϕ (= 0° and 45°) by using the laser etching technique. As a result, J_c at $\phi = 0^\circ$ was higher than one at $\phi = 45^{\circ}$ in the films with BTbO contents of 12.0 and 20.0 vol.% at 77 K and at fields of 5 T or more. The anisotropy of J_c in high fields for current directions was possibly explained by the rectangular shape of the BTbO nanorods.

Index Terms—Artificial pinning centers, BaTbO₃, critical current density, high fields, high temperature superconductor, inplane anisotropy, nanorod, SmBa₂Cu₃O_y.

I. INTRODUCTION

RARE-EARTH cuprate superconductors $REBa_2Cu_3O_y$ (REBCO, RE = Sm, Y, etc.) have a higher critical temperature T_c than liquid nitrogen temperature, in contrast to the conventional low-temperature superconductors. Thus, it is possible to operate REBCO coated conductors at 77 K using liquid nitrogen. In addition, their critical current density J_c is high under considerable applied fields that are suitable for transport, electric, and medical applications such as maglev [1], SMES [2], and MRI [3]. However, it has a disadvantage that its J_c decreases at a high temperature and a high magnetic field compared with LTS.

Various kinds of artificial pinning centers (APCs) have been studied to improve in fields J_c [4] [5]. In particular, it is very effective to introduce BaMO₃ (BMO: M = Zr, Hf, Sn, etc.) into the REBCO films. BMO has the same perovskite structure as REBCO and self-organizes into nanorods in the films. BaHfO₃ (BHO) [6] and BaZrO₃ (BZO) [7] are mainly used as BMO nanorod materials and their doping into the REBCO films enhances J_c in fields a few times larger than non-doped films. However, T_c of REBCO films with a large content of BHO or BSO is greatly degraded [8] [9] [10].

BaTbO₃ (BTbO) is one of BMO materials for REBCO films [11]. R. Kita et al. [12] reported that T_c keeps 90 K or more for a high doping level of BTbO such as 20.0 vol.% in YBa₂Cu₃O_y bulk. It is also reported that T_c is robust for a high doping level of BTbO in SmBCO films and BTbO formed thick nanorods in SmBCO with a large amount of BTbO [13]. From our previous research with the transmission electric microscopy (TEM) technique, the cross-section of BTbO nanorods tends to be rectangular. We considered that the rectangular nanorods would produce an in-plane anisotropy for the flux pinning. In this study, we fabricated SmBCO films with various BTbO contents and investigated their microstructures and in-plane anisotropy of their transport properties. To clarify the in-plane anisotropy of the flux pinning by BTbO thick rectangular nanorods, we compared the above results with the in-plane anisotropy in the pure SmBCO and the BHO-doped SmBCO films.

II. EXPERIMENTAL

SmBCO thin films with BHO or BTbO were deposited by the pulsed laser deposition (PLD) method on CeO₂ buffered IBAD-MgO metallic substrates. A modified target method was used to introduce BTbO and a mixed target method was used for the introduction of BHO. The BTbO contents were up to 20.0 vol.% and the BHO content was 2.5 vol.%. The deposition

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TABLE I DEPOSITION CONDITIONS OF THE PURE, BHO-DOPED, AND BTBO-DOPED SMBCO FILMS

	pure	BHO-doped	BTbO-doped
Laser Type	KrF-excimer laser		
(wave length)	$(\lambda = 248 \text{ nm})$		
Method	Pulsed Laser Deposition (PLD)		
Target	SmBa ₂ Cu ₃ O _y	SmBa ₂ Cu ₃ O _y	SmBa ₂ Cu ₃ O _y
U	,	+ BaHfO ₃	+ BaTbO ₃
Substrate	CeO2 buffered IBAD-MgO		
Energy density	1.0 J/cm^2		
Target-substrate	60 mm		
distance			
Substrate tem-	860°C	860°C	880°C
perature			
Partial O ₂	400 mTorr		
pressure			
Thickness	200-300 nm		
Doping method	Non	Mixed Target	Modified Target
BMO content	0 vol.%	2.5 vol.%	6.8-20.0 vol.%

conditions are shown in Table 1. The deposited films were patterned by laser etching to prepare the microbridge shape with a strip dimension of 100 µm wide and 1 mm long for measurement. To fix the directions of current in the films, the microbridges were fabricated with different ϕ (= 0° and 45°) as shown in Fig. 1. H. Safar *et al.* [14] called these microbridges as "*I*" and "*Z*" shapes. The ϕ is defined as an in-plane angle from the *a* or *b*-axes of SmBCO. The four-probe method was used for the measurement of T_c and J_c . The field dependence of J_c of films was measured at 77 K, at 0-9 T, and $\phi = 0^\circ$ and 45°. The angular dependence of J_c was also measured at 77 K and 1 T, with varying magnetic field orientation angle. J_c at B//c was defined by 1 µV cm⁻¹ criterion. The formation of BTbO nanorods in the films was observed by using the TEM technique.



Fig. 1. Schematic drawings of microbridges in the SmBCO films with $\phi = 0^{\circ}$ and 45°, where ϕ was defined as an in-plane current angle from the *a* or *b*-axes of SmBCO.

III. RESULTS AND DISCUSSION

Fig. 2 shows Ba MO_3 (BMO: M = Tb, (Sm, Nb), Zr, Hf, Sn) content dependence of T_c in BMO-doped SmBCO films for various kinds of M [15, 16]. T_c decreased below 90 K for 10 vol.%

doping of BHO or BZO or more. On the other hand, T_c keeps 90 K or more for a high doping level of BTbO or Ba₂SmNbO₆ (BSNO) such as 20 vol.% in SmBCO films. In particular, T_c of the 34.7 vol.% BTbO-doped film was 91.1 K. One of the reasons for this would be that the nanorods number density of BTbO was lower than one of BHO and BZO [16]. Since BTbO formed thicker nanorods than BHO and BZO, the number density of BTbO was lower than one of BHO and BZO. In that case, the lattice stress in the SmBCO matrix introduced by BTbO was smaller than one by BHO or BZO.



BMO content [vol%]

Fig. 2. $BaMO_3$ (BMO: M = Tb, (Sm, Nb) [15], Zr [16], Hf [16], Sn [16]) content dependence of critical temperatures in BMO-doped SmBCO films.



Fig. 3. Magnetic field angle dependence of J_c in the BTbO-doped SmBCO films at 77 K and 1 T.

Fig. 3 shows the magnetic field angle dependence of J_c of the pure and BTbO-doped films at 77 K and 1 T. J_c at B//c was lower than one at B//c in the pure SmBCO films because of intrinsic pinning and stacking faults [17]. J_c at B//c was higher than one at B//ab in the 9.0-20.0 vol.% BTbO-doped SmBCO films. This indicated that *c*-axis correlated APCs were introduced into the films. In addition, J_c at 77 K and 1 T was almost the same in the 12.0-20.0 vol.% BTbO-doped films. From the TEM images of Fig. 5, BTbO formed nanorods. This is a point to which we shall return later. On the other hand, J_c at B//c was lower than one at B//ab in the 6.8 vol%. Tsuchiya *et al.* [13]

reported that BTbO formed *c*-axis correlated APCs in SmBCO films only with a large amount of BTbO.

Figs. 4 (a), (b) shows the magnetic field dependence of J_c of the pure, BHO-doped, and BTbO-doped samples at a temperature of 77 K, fields of 0-9 T, and current directions of $\phi = 0^{\circ}$ and 45°. As a result, J_c at $\phi = 0^{\circ}$ was higher than the one at $\phi = 45^{\circ}$ in the thin films with BTbO contents of 12.0 and 20.0 vol.% at 77 K and 5 T or more. On the other hand, J_c is isotropic in the



Fig. 4. Magnetic field dependence of (a) J_c in the pure, BHO-doped , and the 6.8 vol.% BTbO-doped SmBCO films, (b) J_c in the BTbO-doped SmBCO films, and (c) normalized anisotropy of J_c in those SmBCO films at 77 K, B//c, $\phi = 0^{\circ}$ and 45°.

pure, 2.5 vol.% BHO-doped, and 6.8 vol.% BTbO-doped SmBCO films. Fig. 4 (c) shows the magnetic field dependence of normalized anisotropy of J_c in the SmBCO films with BHO or BTbO at 77 K, B//c, $\phi = 0^{\circ}$ and 45°. The anisotropy showed a ratio of J_c between 0° and 45°, which was normalized by the one at 0 T. As a result, the anisotropy was 0.95-1.65 in the pure, 2.5 vol.% BHO-doped, and 6.8 vol.% BTbO-doped SmBCO films at 77 K and at 9 T or less. However, the anisotropy exceeded 3 in the 12.0 and 20.0 vol.% BTbO-doped samples at 77 K and at fields more than 7 T. In particular, an anisotropy of 13 times was obtained in the 12.0 vol.% doped film at 77 K and 9 T as the largest value in this study.



Fig. 5. Plan-view TEM images of the (a) 12.0 vol.% and (b) 20.0 vol.% BTbO-doped SmBCO films. The inset shows cross-sectional TEM images of those films.

Fig. 5 shows plan-view and cross-sectional TEM images of the (a) 12.0 vol.% and (b) 20.0 vol.% BTbO-doped films. The white arrows in the planar TEM images indicate BTbO rectangular nanorods. From the TEM images, BTbO had a shape of thick nanorods with diameters of ~20 nm in the 12.0 vol.% and 20.0 vol.% doped films. The matching fields B_{Φ} of the 12.0 vol.% and 20.0 vol.% doped films were 1.5 T and 1.1 T calculated by the nanorods densities from the planar TEM images, respectively. Moreover, planar the TEM images showed that BTbO formed rectangular nanorods. The sides of the rectangles were parallel to the a or b-axes of SmBCO. For the 12.0 vol.% and 20.0 vol.% BTbO-doped films, 37-39.0% of the BTbO nanorods are identified to be rectangular. In addition, the averages and standard deviations of misalignment angles between the rectangular nanorods and the a or b-axes of SmBCO were less than 1° and 13-16° for both the films, respectively. Therefore, the rectangular nanorods were slightly aligned along a or b-axes of SmBCO.

According to the results, we now proceed to discuss four possible origins for the anisotropic J_c in BTbO-doped films. The first possibility is potential from pinned vortices. With Ginzburg-Landau parameter of $\kappa \cong 3$, the saturation number of vortices of a nanorod N_s is estimated as $N_s \cong d/2\xi(T)$ [17], where the maximum number of vortices are pinned at the nanorods. N_s was 3.6 in a case of those BTbO nanorods with diameters of ~20 nm at 77 K. As shown in Figs. 6 (a) (b), about 4 vortices are pinned by nanorods at fields more than 5 T (= $N_s B_{\Phi}$). There are sides of the rectangles of nanorods vertical to the Lorenz force at $\phi = 0^\circ$, so vortices pinned by nanorods prevent vortices

around nanorods from flowing. On the other hand, the angle between the sides and the Lorenz force is at $\phi = 45^\circ$, so vortices around nanorods easily flow. Therefore, J_c at $\phi = 0^\circ$ is higher than one at $\phi = 45^\circ$ at high fields. This origin is negative because vortices pinned by nanorods should move at 5 T which was



Fig. 6. Schematic drawings of (a) (b) pinned or flowing vortices around BTbO rectangular nanorods and (c) (d) BTbO rectangular nanorods and flux penetration in the BTbO-doped SmBCO films with (a) (c) $\phi = 0^{\circ}$ and (b) (d) 45° at fields more than 5 T (*B*//*c*).

higher than $N_{\rm s}B_{\Phi}$.

The second possibility is current crowding [18]. Pinned vortices produce electric fields around the nanorods. Since the electric fields are higher at the edges of rectangular nanorods higher than ones at the sides, vortices must be easily pinned and depinned by nanorods at the edges as shown in Figs. 6 (c) (d). When ϕ is 0°, the Lorenz force is parallel or vertical to the sides of nanorods. Thus, J_c at $\phi = 0^\circ$ is higher than one at $\phi = 45^\circ$. It corresponds with the experimental results.

The third possibility is the effect of twin boundary [19]. H. Safar *et al.* [14] reported that J_c in fields could be increased by producing conductors with twin boundaries tiled 45° with respect to the current flow. J_c in the pure and BHO-doped SmBCO films must be anisotropic if twin boundaries produced the anisotropy of J_c in the BTbO-doped sample.

The fourth possibility is vortex channeling. M. Pannetier *et al.* [20] reported an anisotropy of the flux penetration in $YBa_2Cu_3O_y$ films containing a square array of antidots. In the antidots array, vortices easily penetrate along the direction between the nearest antidots or parallel to the square lattice, which leads the anisotropy of vortices penetration. In this model, the saturation number of vortices in a hole must be equal to 1 [17] [21]. Therefore, this model cannot be applied to the BTbO-doped films in this study.

From the above consideration, we insist that current crowding probably leads the anisotropy of J_c in the BTbO-doped films. It was found that it is important to consider the orientation of a buffer layer in case of introducing nanorods into REBCO films from this study. It is an advantage of BTbO that it has a wide window for doping. In addition, J_c possibly increases more than 10 times in high fields by considering the current direction. There is a possibility that REBCO films thin films would be used for high field applications if thin rectangular nanorods whose sides are aligned can be introduced into REBCO films.

IV. CONCLUSION

We fabricated the pure, BHO-doped, and BTbO-doped SmBCO films on IBAD-MgO buffered metal substrates by using PLD methods. We introduced BHO by using a mixed target and BTbO by using modified targets. We measured the anisotropy of transport properties by making microbridges with different ϕ (= 0° and 45°). T_c was 90 K or more in the SmBCO films with a high content of BTbO such as 20.0 vol.%. Field dependence of J_c was measured at 77 K and 1 T. J_c at B//ab was higher than one at B//c in the 0 and 6.8 vol.% BTbO-doped films. In contrast, J_c at B//c was higher than one at B//ab in the 12.0-20.0 vol.% BTbO-doped samples. This indicates that BTbO formed nanorods parallel to *c*-axis of SmBCO with a large amount of BTbO. To clarify the in-plane anisotropy, we measured J_c at 77 K, 0-9 T, and $\phi = 0^\circ$ and 45°. As a result, J_c was isotropic in the pure, BHO-doped, and 6.8 vol.% BTbOdoped SmBCO films. On the other hand, J_c at $\phi = 0^\circ$ was higher than one at $\phi = 45^{\circ}$ in the films with BTbO content of 12.0 and 20.0 vol.% at 77 K and 5 T or more. In particular, an anisotropy reached 10 times or more in the 12.0 vol.% doped film at 77 K and 9 T. TEM images showed that the shapes of the BTbO-APCs were rectangular nanorods and B_{Φ} was 1.1-1.5 T in the 12.0 and 20.0 vol.% BTbO-doped samples. The difference of J_c was explained by an anisotropy of flux penetration by current crowding. The shapes of the rectangular BTbO nanorods raised electric fields at the edges of the rectangles higher than ones at the sides. The higher electric fields made vortices pin and depin easily by nanorods at the edges. So J_c at $\phi = 0^\circ$, when the Lorenz force was parallel or vertical to the sides of the rectangles, was higher than one at $\phi = 45^{\circ}$, when the angle between the Lorenz force and the side was 45°.

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