## Tilt angle dependences of vortex structure and critical current density at low-angle grain boundaries in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> films

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A tilt angle dependence of vortex structure was studied at low-angle grain boundaries (LAGBs) in  $YBa_2Cu_3O_{7-x}$  films. The 2° tilt LAGB did not degrade  $J_c$  of the film in a self-field, demonstrating the existence of Abrikosov vortices at the 2° LAGB. The magnetic field dependence of the flux flow resistance indicated the existence of Abrikosov Josephson vortices at the 5° tilt LAGB. At the 10° tilt LAGB, a Josephson junction behavior was observed in a self-field, indicating the existence of Josephson vortices. The influence of the LAGBs on critical current density was understood based on these vortices.

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Quantized vortices in high temperature superconductors (HTSCs) exhibit a characteristic behavior at grain boundaries (GBs). Therefore, the GBs in HTSCs are one of the most important systems in vortex physics. Current density-voltage (J-V) curves and critical current density-magnetic field  $(J_c-B)$  characteristics are strongly dependent on the structure of the dominant vortices,<sup>1,2</sup> and three types of vortices have been discussed in superconductors: Abrikosov (A) vortex, Abrikosov–Josephson (AJ) vortex, and Josephson (J) vortex. Abrikosov vortices with a normal core [radius=coherence length  $(\xi)$  generate rich vortex phases below the upper critical field  $B_{c2}[=\phi_0/(2\pi\xi^2)]$ .<sup>1</sup> Based on the vortex phase diagrams, J-V curves and  $J_c-B$  characteristics have been understood in single crystalline HTSCs. In typical Josephson junctions (JJs), J vortices without a normal core exist, and a JJ behavior induces an oscillation in  $J_c$ -B characteristics.<sup>2,3</sup> In addition, Gurevich suggested the existence of intermediate AJ vortices with a J core [radius=characteristic length (l)] at highly transparent weak links.<sup>4,5</sup> The behavior of AJ vortices was experimentally studied at low-angle grain boundaries (LAGBs) in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> (YBCO) films.<sup>6,7</sup> The influence of GBs on the transport properties should be understood based on these vortices.<sup>8</sup> However, although numerous researchers have measured  $J_c$  and J-V curves in HTSC films with a single GB,8,9 such experimental results have not been systematically discussed based on A, AJ, and J vortices. In this Brief Report, we report a tilt angle dependence of vortex structure at LAGBs in YBCO films. To analyze the structure of the GB vortices, we measured J-V curves and  $J_c$  at LAGBs as a function of tilt angle. Based on the vortex structure, we discuss the influence of LAGBs on  $J_c$  and J-Vcurves.

C-axis-oriented YBCO films, which were 350-390 nm thick, were prepared on single crystal SrTiO<sub>3</sub> (STO) and symmetrical [001] tilt bicrystal STO substrates (tilt angle  $=2^{\circ}, 5^{\circ}, 10^{\circ}$ ) using pulsed laser deposition (PLD). The LAGBs in the YBCO films deposited on polycrystalline metallic substrates using PLD did not meander through the thickness of the films.<sup>10</sup> Therefore, in the present study, we can regard the LAGBs as two-dimensional planar walls, which consist of an array of edge dislocations. According to Frank's formula, the spacing between the dislocations is 11.0, 4.4, and 2.2 nm at 2°, 5°, and 10° tilt LAGBs, respectively.<sup>8</sup>  $J_c$  and J-V curves in a magnetic field parallel to the c axis of the films were measured in a temperature range of 60-77 K. The current was always perpendicular to the LAGB in the BC YBCO films. The YBCO films were etched using the typical lithography technique to form  $60-85 \ \mu m$ wide bridges.  $J_c$  was given by  $I_c/A$ , where  $I_c$  is the critical current (1  $\mu$ V/cm criterion) and A is the cross-sectional area of the bridge. YBCO films on a single crystal substrate and an X° bicrystal substrate are termed as "SC YBCO film" and "X° BC YBCO film," respectively.  $J_{SC}$  and  $J_{BC}$  denote the measured  $J_c$  in SC YBCO and BC YBCO films, respectively. In addition,  $J_{GB}$  and  $J_{IG}$  are defined as the  $J_c$  determined at LAGB and the  $J_c$  governed by the vortex pinning in grains, respectively.<sup>11</sup>

 $T_c$  values in the SC YBCO film and the 2°, 5°, and 10° BC YBCO films were 89.6, 89.5, 89.3, and 88.6 K, respectively. The self-field  $J_c$  values in the grains, which were obtained from the  $J_c$  measurements in the SC YBCO films, were 1.1–1.4 MA/cm<sup>2</sup> at 77 K. Figure 1(a) shows  $J_{BC}$  (0 T)/ $J_{SC}$  (0 T) as a function of tilt angle of LAGB at 77 K.



FIG. 1. (a) A tilt angle dependence of  $J_{BC}$  (0 T)/ $J_{SC}$  (0 T) at 77 K in the present study, compared with those at 4 K (Ref. 9) and 77 K (Ref. 12) in previous reports. (b) Magnetic field dependences of  $J_{SC}$  and  $J_{BC}$  in the 2°, 5°, and 10° BC YBCO films at 77 K.

The  $J_{BC}$  (0 T)/ $J_{SC}$  (0 T) decreased with increasing tilt angle after initially exhibiting a plateau at 0°–2° in the present study, as was observed in previous reports.<sup>9,12</sup> The slight difference in the  $J_{BC}$  (0 T)/ $J_{SC}$  (0 T) behavior between the present study and the previous reports is due to the difference in the carrier-doping state at the LAGBs. Figure 1(b) shows magnetic field dependences of  $J_c$  at 77 K as a function of tilt angle in a magnetic field parallel to the *c* axis. Verebelyi *et al.* obtained a similar result.<sup>13</sup>

The  $J_{BC}$  in the 2° BC YBCO film is almost the same as the  $J_{SC}$  in the entire magnetic field. The  $J_{GB}$  for the 2° LAGB is determined by depinning because the tilt angle is sufficiently small. In the SC YBCO films, the vortex pinning is mainly due to threading dislocations with a density of  $10-100 \ \mu m^{-2}$ .<sup>14</sup> In the vicinity of the crossover from A vortices to AJ ones, the size of the vortex core is not so large  $(\sim\xi)$  even for AJ vortices. Therefore, here, it is considered that the dominant pinning centers at the LAGBs are also the GB dislocations<sup>15</sup> since the crossover from A vortices to AJ ones is discussed. Because the  $J_c$  in a very low field is determined by the single vortex pinning,<sup>1,14</sup> the  $J_c$  in the grains and the  $J_c$  at the 2° tilt LAGB in a self-field are thought to be determined by the single vortex pinning due to the dislocations. The  $J_c$  in the single vortex pinning region is given by  $U_p/x\phi_0$ , where  $U_p$  is the pinning potential and x is the radius of the vortex core. When A vortices exist at the LAGB, x =  $\xi$  both at the LAGB and in the grains. In this case, since the  $(U_p/x\phi_0)$  value at the LAGB is the same as that in the grains,  $J_{BC} = (J_{GB} \text{ for A vortices}) = J_{IG} = J_{SC}$  in the single vortex pinning region in the BC film. On the other hand, the xvalue for AJ vortices is larger than that for A vortices. This means that the  $(U_p/x\phi_0)$  value for AJ vortices is smaller than that for A vortices. Thus, as A vortices turn into AJ vortices at the LAGB,  $J_{BC}$  starts to be determined by the behavior of AJ vortices at the LAGB and becomes smaller than  $J_{SC}$  $[J_{BC}=(J_{GB} \text{ for AJ vortices}) < (J_{GB} \text{ for A vortices}) = J_{IG}=J_{SC}]$ in the single vortex pinning region. The nearly identical  $J_c$ values for the SC YBCO film and the 2° BC YBCO film in a self-field indicate that A vortices existed at the 2° tilt LAGB. On the other hand, if the  $J_{BC}$  in a high field was determined by the depinning of the A vortices at the 2° tilt LAGB, the  $J_{BC}$ -B curve should deviate upward from the  $J_{SC}$ -B curve near the irreversibility field  $(B_{irr})$  (Ref. 1) because the dislocation density at the 2° tilt LAGB is much higher than that in



FIG. 2. Magnetic field dependences of *J*-*V* curves at 77 K in the (a) 5° BC YBCO film and the (b) 10° BC YBCO film. (c) and (d) are enlarged views of (a) and (b). (e) A magnetic field dependence of  $R_FA$  at 77 K in the 5° BC YBCO film. Open squares denote the measured data. A solid line shows  $R_FA=2 \times 10^{-10} (B/B+0.13)^{1/2}$ . (f) A magnetic field dependence of (linear resistance × area) at 77 K in the 10° BC YBCO film. The linear resistance in (f) does not correspond to  $R_F$ . Open circles denote the measured data. A solid line is a guide to the eye. Our previous paper originally described the main parts of (a), (c), and (e) (Ref. 7). Here, the comparison of the results in the 5° BC YBCO film with those in the 10° BC YBCO film is important.

the grains. However, the behavior of the  $J_{BC}$ -B curve in the 2° BC YBCO film is similar to that of the  $J_{SC}$ -B curve, suggesting that the  $J_{BC}$ -B curve of the 2° BC YBCO film in the high field was determined by the depinning in the grains.

 $J_{BC}$  is smaller than  $J_{SC}$  in a self-field in the 5° and 10° BC YBCO films, showing that the GB vortices at the 5° and 10° tilt LAGBs were not A vortices. Figure 2 shows *J*-*V* curves for  $B \parallel c$  at 77 K in the 5° BC YBCO film [(a) and (c)] and those in the 10° BC YBCO film [(b) and (d)]. At  $B^*$  (~0.25 T), the curvature of the *J*-*V* curves in the 5° BC YBCO film changed. The *J*-*V* curves have a positive curvature (convex downward) below 0.2 T, but those in magnetic fields above 0.3 T have a negative curvature (convex upward). This behavior also seemed to be present for other conditions and other films.<sup>7,16</sup> The  $R_F$ -B curve in Fig. 2(e) indicates that the *J*-*V* curves with a positive curvature were determined by the flux flow (FF) of AJ vortices. However, as discussed in our previous paper,<sup>7</sup> the decrease in  $J_{GB}$  with

increase in *B* and the negative curvature of *J*-*V* curves for  $B^* < B < B_{cr}$  cannot be simultaneously explained by the depinning and FF of AJ vortices. Thus, the  $J_{GB}$  was determined by the depinning of AJ vortices in magnetic fields lower than  $B^*$ , but it was governed by a JJ behavior in magnetic fields higher than  $B^*$  in the 5° LAGB. *l* values for AJ vortices can be obtained from the magnetic field dependence of flux flow resistance,  $R_F$ .<sup>6.7</sup> Theoretically,  $R_F$  is given by

$$R_F = R_0 \sqrt{\frac{B}{B + B_0}} \tag{1}$$

for low current density, where  $B_0 = \phi_0 / (2\pi l)^2$  and  $R_0$  is a proportionality constant.<sup>5,6</sup> Figure 2(e) shows the  $R_FA$ -B curve in the 5° BC YBCO film at 77 K, which was obtained from dV/dJ in the linear resistance region in magnetic fields lower than 0.2 T. By fitting the measured  $R_FA$ -B curve using Eq. (1),  $B_0=0.13$  T and l=20 nm were obtained. This suggests that AJ vortices ( $l=20 \text{ nm} \ge \xi$ ) existed at the 5° LAGB at 77 K.  $(\phi_0/B)^{1/2}$  was proportional to *l* regardless of temperature in our previous study,<sup>7</sup> showing that the change of the curvature of J-V curves in the magnetic field was induced by the overlap of J cores in AJ vortices. The overlapping field  $B^*$  is given by  $\phi_0/(4.5l)^2$  since l=20 nm and  $B^*$  $\sim 0.25$  T at 77 K. The overlap of J cores in AJ vortices was observed also at antiphase boundaries, although the difference in boundary structure varies the behavior of the  $J_c$ -B curves.<sup>17</sup> On the other hand, similar magnetic field dependences of  $J_{BC}$  and  $J_{SC}$  were observed in the 5° BC YBCO film in magnetic fields higher than 4.5 T. In addition, nonlinear J-V curves were observed at 5 and 7 T. These results indicate that the  $J_{BC}$  was determined by  $J_{IG}$  in a magnetic field higher than  $B_{cr}^{LC}$  (=4.5 T).<sup>11,18</sup> Here,  $B_{cr}$  is defined as the magnetic field where the behavior of  $J_{BC}$  starts to agree with that of  $J_{SC}$ .<sup>18</sup>

The *J*-*V* curves in the 10° BC YBCO film have a negative curvature even in a self-field. In addition, the linear resistance of the *J*-*V* curves is not dependent on magnetic fields, as shown in Fig. 2(f). These results are similar to the behavior of *J*-*V* curves in the typical JJs where J vortices exist. Thus, at the 10° tilt LAGB, J vortices existed and JJ behavior determined the  $J_{GB}$  regardless of magnetic fields. In the typical JJs, the Josephson penetration depth  $\lambda_J$  is obtained from the  $J_c$  value at 0 T. As shown in Fig. 3, the relationship of  $\lambda_J > \lambda$  proves the existence of J vortices at the 10° LAGB, where  $\lambda$  is the London penetration depth. On the other hand, nonlinear *J*-*V* curves and similar field dependences of  $J_{BC}$  and  $J_{SC}$  in the high fields show that the  $J_{BC}$  in the 10° BC YBCO film was determined by  $J_{IG}$  at a magnetic field higher than  $B_{cr}$  (=6 T).

The GBs where AJ or J vortices exist should be a planar weak-superconducting barrier. Therefore, the crossover from A vortices to AJ ones results from the overlap of the nonsuperconducting cores around the edge dislocations at the LAGB. The overlapping angle of  $3^{\circ}-5^{\circ}$  has been obtained from a theoretical calculation by Gurevich and Pashitskii.<sup>19</sup> The elongation of the vortex core due to the crossover from A vortices to AJ ones induces the deviation of  $J_{BC}/J_{SC}$  from one in a self-field.



FIG. 3. Temperature dependences of the  $J_{BC}$  in a self-field, which were measured in the present study and in previous reports (Refs. 9, 20, and 21). Open and solid symbols denote the  $J_{BC}$  values, which were determined by FF and JJ behaviors in a self-field, respectively. A solid line denotes the calculated  $0.4J_b^*$  curve.

To study the crossover from AJ vortices to J ones between 5° and 10°, we discuss the  $J_{BC}$  in a self-field. When *l* becomes larger than  $\lambda$ , AJ vortices turn into J vortices. The GB depairing current density for AJ vortices,  $J_b$  at  $l=\lambda$  is given by  $J_b^*$  (Ref. 5)

$$J_b^* = \frac{\phi_0}{4\pi\mu_0\lambda^3}.$$
 (2)

We can obtain Eq. (2) also from the GB depairing current density of the typical JJs for  $\lambda_J = \lambda$ . Figure 3 shows temperature dependences of  $J_{BC}$  in a self-field, which were obtained in the present study and in previous reports.<sup>9,20,21</sup> The calcu- $\phi_0 = 2.07 \times 10^{-15}$  Wb, curve where lated  $0.4J_{h}^{*}$  $\lambda = \lambda_0 \{1 - (T/\tilde{T}_c)^4\}^{-1/2}, \lambda_0 = 150 \text{ nm}, \text{ and } T_c = 90 \text{ K} \text{ is also}$ shown in Fig. 3. In Fig. 3, the FF and JJ regions in a selffield are observed for high  $J_{BC}$  and for low  $J_{BC}$ , respectively, and the calculated  $0.4J_b^*$  curve defines the boundary between them so well regardless of temperature and samples. Since the depinning  $J_c$  in the FF region is smaller than the GB depairing current density,<sup>6,7</sup> Fig. 3 suggests that (GB depairing current density) > (depinning  $J_{GB}$ ) =(measured  $J_{GB}$ )>0.4 $J_{b}^{*}$  in a self-field in the FF region. This shows  $l < \lambda$  in the FF region. On the other hand, Fig. 3 shows (GB depairing current density) = (measured  $J_{GB}$ ) < 0.4 $J_{b}^{*}$  in a self-field in the JJ region. These results strongly suggest that the crossover in J-V curves from FF to JJ behavior in a self-field is induced by the change of vortex structure from AJ vortices to J ones. The prefactor of 0.4 is plausible due to the reduced transmission area of the LAGB. Figure 3 indicates that the supercurrent flows through 40% of the transparent area at the LAGB. The density of facets at LAGBs is too  $low^{22}$  to explain the prefactor of 0.4. In addition, it is expected that the meandering of LAGBs due to the facets may increase the effective current flowing area from that of the straight LAGBs. Therefore, the prefactor of 0.4 should be explained by the GB dislocations,<sup>12</sup> not by the facets. The LAGB consists of highly distorted regions (dislocation core) and elastically strained regions. The crossover from FF to JJ behavior in a self-field has been observed between 8° and 15°, regardless of samples and temperature.<sup>8,9,20</sup> Assuming that the spacing between the dislocations is 2.5 nm and all of



FIG. 4. (Color online) The determination mechanism of  $J_{BC}$  and *J-V* curve and the vortex structure at [001] tilt LAGBs in BC YBCO films. "FF" means that  $J_{BC}$  is determined by depinning.

the current flows through the elastically strained regions, the radius of the highly distorted region is estimated to be 0.8 nm ( $\sim 2b$ , where *b* is the Burgers vector). An electron holography study on the electrostatic potential distribution at a 4° [001] tilt LAGB indicated that the dislocation core potential is in the range of  $\pm 0.85$  nm from the dislocation cores.<sup>23</sup>

Figure 4 summarizes the mechanism of  $J_{BC}$  and J-V curve and the vortex structure at [001] tilt LAGBs in BC YBCO films. When AJ vortices existed at the LAGB, the crossover from FF to JJ behavior occurred at  $B^*[\sim \phi_0/(4.5l)^2]$ . As lapproaches  $\xi(AJ \rightarrow A)$ ,  $B^*$  becomes  $\phi_0/(4.5\xi)^2$ , which is comparable to  $B_{c2}$ . On the other hand, as l approaches  $\lambda(AJ \rightarrow J)$ ,  $B^*$  becomes  $\phi_0/(4.5\lambda)^2$ , which agrees well with the  $B_{c1}$  in the typical JJs  $[B_{c1}=\phi_0/(\pi^2\lambda_J\lambda)]$  for  $\lambda_J=\lambda$ . Thus,  $B_{c2}$  for A vortices and  $B_{c1}$  for J vortices are continuously linked via  $B^*$  for AJ vortices.

Since  $J_{BC}$  is determined at the preferential voltage generation portion in the films,  $J_{BC} = J_{GB}$  for  $B < B_{cr}$  and  $J_{BC} = J_{IG}$ for  $B > B_{cr}^{-11}$  When A vortices exist at a LAGB, the local  $B_{irr}$ at the LAGB is determined by the thermal fluctuation and pinning of the A vortices. Since the  $B_{irr}$  is a function of the density of c-axis correlated pinning centers,<sup>1</sup> the high density of GB dislocations can enhance the  $J_{GB}$  for the A vortices at the [001] tilt LAGB by increasing the local  $B_{irr}$  at the LAGB. On the other hand, since a JJ behavior is not affected by the thermal fluctuation so strongly, the  $J_{GB}$ , which is determined by a JJ behavior (GB vortex: AJ or J vortex), remains nonzero even at the local  $B_{irr}$  for the A vortices in the grains. Thus,  $J_{GB}$  is higher than  $J_{IG}$  in the high field regardless of the GB vortex structure, although the mechanism of the high  $J_{GB}$ is dependent on the GB vortex structure. Since  $J_{GB}$  decreases with increasing tilt angle,  $B_{cr}$ , which is determined by the relation of  $J_{GB}(B_{cr}) = J_{IG}(B_{cr})$ ,<sup>11</sup> increases with increase in tilt angle.<sup>18</sup>

In conclusion, A vortices, AJ vortices, and J vortices systematically appeared at the LAGBs in the YBCO films depending on the tilt angle. The 2° tilt LAGB did not reduce  $J_c$  of the BC YBCO film in a self-field, demonstrating the existence of A vortices at the 2° tilt LAGB. The mechanism of the *J*-*V* curve was changed at the magnetic field in the 5° tilt LAGB where the AJ vortices existed. Due to the J vortices at the 10° tilt LAGB, the 10° BC YBCO film exhibited a JJ behavior even in a self-field. The influence of LAGBs on  $J_c$  and *J*-*V* curves was explained based on A, AJ, and J vortices.

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