# Critical current density of $YBa_2Cu_3O_{7-\delta}$ low-angle grain boundaries in self-field

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A study has been perfomed on the superconducting critical current density $J_c$ flowing across low-angle grain boundaries in epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$. The materials studied were dual grain boundary rings deposited on $SrTiO_3$ and containing 2°, 3°, 5°, and 7° tilt boundaries. The current density in self-field was determined by magnetometric methods at temperatures from 5 K to $T_c$. We conclude that at the higher temperatures of coated conductor applications, there is limited potential for improving $J_c$ by reducing the grain boundary angle below $\sim 3°$. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1360230]

Well-aligned grains are necessary for optimum critical current density $J_c$ in high-temperature superconductors. A large misalignment of adjacent grains suppresses the order parameter at the grain boundary (GB) and results in Josephson tunneling behavior[1] and a near-exponential decrease in $J_c$ with misorientation angle.[2–4] In the case of $YBa_2Cu_3O_{7-\delta}$ (YBCO) low-angle grain boundaries ($\theta \lesssim 4°$), the boundary is comprised of a periodic array of dislocation cores separated by only slightly perturbed material[5] providing a strong conduction channel wider than the in-plane coherence length. This low-angle regime is particularly important to understanding transport in the highly-textured coated conductors. Both of the most prominent coated conductor methodologies, rolling-assisted biaxially textured substrate[6,7] and ion-beam assisted deposition,[8] can provide this level of texture. Our previous transport results concluded that a 2° GB was inherently different from a 5° GB due to the weak-linked characteristics of the 5° GB.[9] In that study, grain-like voltage–current relations were observed, and no reduction in $J_c$ could be measured across the 2° GB compared with its adjacent grains. However, those results were regarded as inconclusive since the estimated grain boundary voltages were less than those of the grain for reasonable sample geometry. Due to the important implications for coated conductors and the fundamentals of epitaxial YBCO films, we have investigated the properties of low-angle GB utilizing a high sensitivity magnetometer technique. From their magnetic moments, we deduce the persistent currents flowing in thin YBCO rings with and without 2°, 3°, 5°, and 7° [001]-tilt boundaries, and compare these results.

Films were prepared by pulsed laser deposition[7] of YBCO on $SrTiO_3$ (STO) bicrystal substrates with a single [001]-tilt boundary. Using standard optical photolithography techniques, rings were produced with an outside diameter of 3 mm and a width of 100 $\mu$m. A ring was patterned across the GB of the bicrystal, resulting in two GB's included in each "GB ring." For comparison, a companion ring was patterned beyond the grain-boundary region of each substrate to provide a control sample with the intragrain properties, which will be referred to as the "grain ring." The nominal thickness of the films was 200 nm.

Magnetic measurements were conducted with a quantum design MPMS-7 magnetometer. The separated, individual ring samples on rectangular STO substrates were attached to Si mounting disks using cellulose nitrate cement and placed into a Mylar tube for support. The magnetic field $H$, applied perpendicular to the plane of the ring, was ramped to 3000 G at 5 K to induce the maximum circulating persistent currents within the ring. The applied field was then reduced to zero, inducing maximum currents in the opposite sense. The resulting magnetic moment was measured while increasing the temperature from 5 to 95 K in 1 K steps. For the grain rings, $J_c$ was calculated using the modified Bean critical state model,

$$J_c = \frac{30 m_{\text{ring}}}{RV - R'V'}, \tag{1}$$

where $m_{\text{ring}}$ is the magnetic moment in units of emu, $V$ is the volume of a cylinder with the outside radius $R$ of the ring, and $V'$ is the volume of the "missing" cylinder with inside radius $R'$. For the GB ring, we determined the current across the boundary from the measured magnetic moment by removing the background signal arising from currents circulating within the "strip" of YBCO but not crossing the GB. (This procedure was verified by measurements on an open circuit ring in which a line was etched across its entire 100 $\mu$m width; from these data, the Bean model, with a different geometrical factor, also yields the grain $J_c$ of a homogeneous YBCO strip.) In Table I, the relative magnitudes of the corrections are illustated by the ratio of the apparent GB $J_c$ [as deduced from the Bean relation Eq. (1)] to the actual $J_c$ of the boundary, all at 5 K.

We consider and discuss some surprising findings on the lowest angle grain boundary (2°), then return to the measurements on the remaining low angle GB materials. The

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TABLE I. Comparison of critical current density at 5 K for YBCO grain boundaries and their companion grains. Data are calculated values of $J_c$ from the maximum magnetic moment of persistent currents in a narrow ring of YBCO on $SrTiO_3$ substrates, in self-field.

| Misorientation (deg) | GB ring $J_c$ (MA/cm$^2$) | Grain ring $J_c$ (MA/cm$^2$) | $(J_c^{GB}/J_c^{Grain})$ | $(J_c^{GB,corr}/J_c^{GB,Bean})$ |
|---|---|---|---|---|
| 1.8 | 35 | 39 | 0.90 | 1 |
| 1.8 | 36 | 37 | 0.97 | 1 |
| 2.8 | 9.1 | 19[a] | 0.48 | 1 |
| 2.8 | 11.5 | 23 | 0.50 | 0.99 |
| 5.1 | 0.95 | 18 | 0.053 | 0.48 |
| 5.1 | 1.4 | 34 | 0.041 | 0.56 |
| 7 | 0.85 | 31 | 0.027 | 0.47 |

[a]Value measured with an ''open circuit'' ring in strip geometry.

present results, obtained by magnetometry of the 2° GB ring, confirmed our previous transport measurements by showing no discernible reduction in $J_c$ when compared with the companion grain ring. Figure 1 shows the temperature dependence of $J_c$ for two sets of samples with 2° GB and their companion grain rings. High resolution x-ray diffraction quantified the exact YBCO misorientations: 1.83° [001] tilt and 0.13° [100] tilt. From the Franck formula, the calculated dislocation spacing for a 2° GB is 11 nm, with a maximum strong channel span of 10 nm approximated from visual inspection of transmission electron micrograph images. This corresponds to a minimum reduction of only 10% in effective GB length, which is within the expected accuracy range of the data considering geometrical and experimental errors. A possible conclusion is that the dislocation cores along the GB are no more than 1 nm in size with respect to suppression of the order parameter.

This simplistic view, however, may be overshadowed by the intrinsic structure of YBCO films on a cubic substrate. The twin structure of YBCO could provide an explanation for undiminished $J_c$ values across a 2° GB. Twin boundaries are formed in YBCO films at the tetragonal-to-orthorhombic structural transition upon cooling in oxygen from a high temperature. Variations in twin structure are dependent on strain,

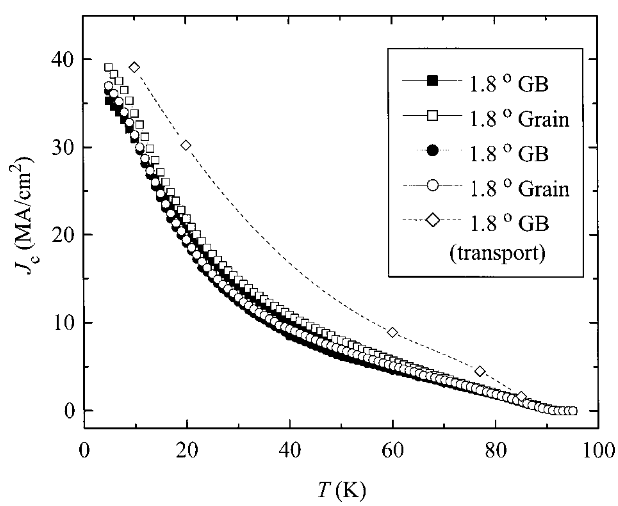


FIG. 1. Magnetometer measurements of persistent current density vs temperature for two 1.8° grain boundary rings and their companion grain rings. Transport data from a 1.8° single grain boundary are shown for comparison.

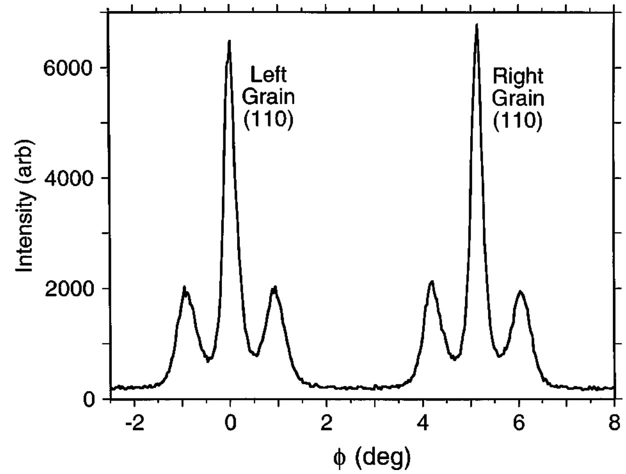


FIG. 2. X-ray $\phi$ scan though the YBCO {225} peak on a 5.1° bicrystal showing the in-plane angular relation between various YBCO domains.

ambient oxygen pressure and cooling rate.[10] Twin boundaries in films are found to be preferentially aligned along the ⟨110⟩ and ⟨1$\bar{1}$0⟩ of lattice-matched cubic substrates. In this case, the orthorhombic nature of the lattice is accommodated by a ~1.8° misalignment of the (1$\bar{1}$0) planes on either side of a (110) twin boundary.[11] Thus, a YBCO film on a $SrTiO_3$(001) single crystal actually consists of domains with four distinct, symmetry-related in-plane orientations. Figure 2 shows an x-ray phi scan through the YBCO (225) peak including both sides of a 5° bicrystal. Each group of three peaks represent one grain of the bicrystal. The strong central peaks at 0° and 5.1° arise from the two twin domains for which the YBCO {110} planes are aligned with the substrate [$HH0$] direction. The difference of 5.1° between these peaks corresponds to the [001]-axis bicrystal tilt misorientation. The smaller peaks located at ±0.9° (as given by [$2\tan^{-1}(b/a)-90°$]) from the central peaks are reflections from the two possible twin domains associated with the twin planes aligned with the [$H\bar{H}0$] substrate direction. This is also true for the adjacent grain across the GB. We note that although the x-ray analysis does not describe the morphology of domains, it does reveal that twinned YBCO films on single crystal substrates are populated with possible GB misorientation angles of 0.9°, 89.1°, and 90°. Thus the YBCO boundaries located above the intersection of a bicrystal substrate with a tilt boundary of $\theta$° are not composed of simply a single well-defined GB of angle $\theta$. Instead, different YBCO domains can impinge with a variety of misorientation angles, including $\theta\pm0.9°$ and $\theta\pm89.1°$. It is important to note that twin boundaries supply no dislocations, while the interface between the suggested twin domains would produce dislocations with an expected 12 nm period. Such a dilute array of dislocations easily accommodates a superconducting coherence length between dislocations, resulting in the typically observed strongly nonlinear, well coupled Abrikosov superconductor. In this view, the addition of a single 2° GB to the matrix would not be expected to provide additional distinguishable dissipation.

Let us now consider more generally the critical current density for grain boundaries with slightly larger angles. In transport studies, prominant tunneling behavior appears to dominate transport chacteristics for a GB angle above

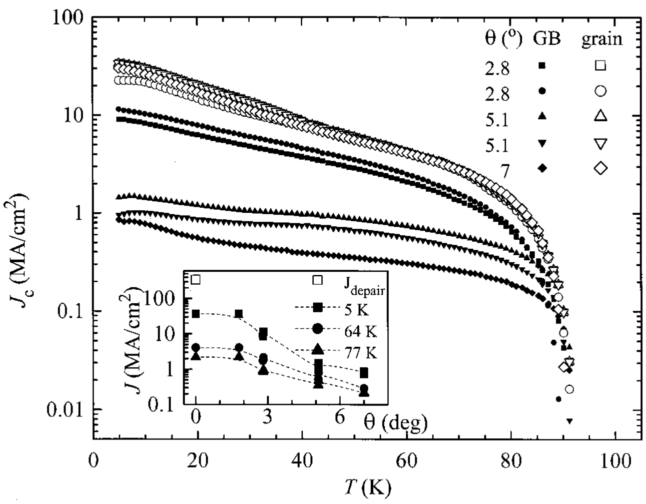


FIG. 3. Magnetometer measurements of critical current density vs temperature for 2.8°, 5.1°, and 7° grain boundary rings and their companion grain rings from the same deposition and substrate. Inset: $J_c$ vs grain-boundary angle at $T=5$, 64, and 77 K, in self-field.

3°–4°, where the grain-like channel between the dislocation cores becomes smaller than a coherence length. The present magnetometry study on GB rings of 2°, 3°, 5°, and 7° provides precise results for the temperature-dependent $J_c$ in the regime of low angle boundaries. Figure 3 shows the temperature dependence of $J_c$ for 3°, 5°, and 7° GB rings, compared to their companion grain rings. Also, Table I lists the $J_c$ values at 5 K for each misorientation angle. The 5° and 7° tilt GBs manifest their tunneling conduction in transport studies via a linear differential $V-J$ characteristic; in these magnetometry studies, a signature of tunneling is a complex in-field $J_c$ with a pronounced dependence on the magnetic field history. In contrast, transport studies of the 2° GB found grain-like, power law characteristics indicative of strong, tunneling-free conduction. The magnetometry studies of both the 2° and 3° rings revealed in-field $J_c$'s with a qualitatively simple, monotonic falloff with $H$ and with minimal-to-no dependence on field history. These results indicate then, a robust current conduction for GBs up to 3°, but pronounced tunneling behavior for GB angles of 5° and above. For the 3° GB, the $J_c$ lies a factor of 2–3 below the grain value, which can be accounted for in part by a reduced cross section of ''good'' material by dislocations.

Qualitatively, Fig. 3 shows a systematic reduction of GB current density with angle. The inset in Fig. 3 shows quantitatively the angle dependence of $J_c$ at temperatures $T=5$, 64, and 77 K. At low temperatures, the $J_c$ rises rapidly as the GB angle decreases in this regime of low angles. For comparison, the (calculated) depairing current density at $T=0$ is included in the inset. At the higher temperatures of coated conductor applications, there is limited potential for improving $J_c$ by reducing the grain boundary angle below ~3°. The present materials, in the temperature range of $LN_2$, already possess the technologically desirable current densities $>10^6$ A/cm$^2$. In this low-angle range, the intragrain $J_c$ becomes the limiting factor, and should become the focus of further improvements, especially in the presence of magnetic fields.

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