Low angle grain boundary transport in $YBa_2Cu_3O_{7-\delta}$ coated conductors

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Second generation, high-temperature superconducting wires are based on buffered, metallic tape substrates of near single crystal texture. Strong alignment of adjacent grains was found to be necessary from previous work that suggested large angle, $YBa_2Cu_3O_{7-\delta}$ [001]-tilt boundaries reduce J_c exponentially with increasing misorientation angle (θ). We pursue the low-θ regime by evaluating single grain boundaries (GB) and biaxially aligned polycrystalline films utilizing both the rolling-assisted biaxially textured substrates and ion-beam assisted deposition coated conductor architectures. Analysis concludes that an exponential dependence on J_c is applicable for $\theta \gtrsim 4°$, where the spacing between the periodic disordered regions along the GB become smaller than a coherence length. © *2000 American Institute of Physics.* [S0003-6951(00)02713-3]

Second generation superconducting wires consist of an epitaxial film deposited on a textured, buffered metallic tape. These ‘‘coated conductors’’ are based on a substrate that is polycrystalline but very well aligned. In fact, grain boundary (GB) misorientations of only a few degrees are typical.[1] This is essential because Chaudhari *et al.*[2] discovered high-angle grain boundaries (HAGB) disrupt the superconducting order parameter, and the transport across the boundary is governed by the Josephson effect. These large grain (GR) misalignments in $YBa_2Cu_3O_{7-\delta}$ (YBCO) result in a perturbed boundary that remains locally ordered and can be described by a specific sequence of structural units.[3] With only small grain misalignment, mismatch is accommodated by periodic dislocations along the GB, separated by only slightly perturbed material. Chisholm and Pennycook[4] investigated the disordered region with transmission electron microscopy (TEM) and correlated atomic displacement to strain which is modeled as a linear decrease in critical current density (J_c) at low θ. Analysis of HAGBs provides a model for the apparent exponential dependence[5] of J_c at large θ due to a linear increase in the width of the boundary.[3] Several studies[5–7] indicate a near-exponential dependence at high θ, but large discrepancies still exist within the literature for the low-angle grain boundary (LAGB) regime. A more complete understanding of LAGBs is necessary for the advanced development of coated conductors targeting power delivery and high magnetic field applications.

First, we address the problem of inconsistent reporting of the ratio J_c^{GB}/J_c^{GR}, which is a widely used quantity based on the assumption that J_c^{GB} scales with J_c^{GR}. In the ideal case this is expected to hold true, but the variables responsible for reduced grain J_c may have a lesser effect on the GB due to its disordered nature. Even an anti-correlation between J_c^{GR} and J_c^{GB} could be possible based on a doping scenario where the preferred stoichiometry of the GB differs from that of the grain. To counter some of these concerns, our approach investigates the grain-GB system through the analysis of the V-J relationship in magnetic field combined with extended V-J curves, which enables simultaneous measurement of both the grain and GB response. This permits a direct comparison of the grain and GB while reducing the chance of experimental variables affecting the results. We combine these measurements with the practice of reporting absolute values of J_c and validating the consistently high-J_c's of the grains with successive patterning.

Two systems have been studied: the first system uses a symmetric $SrTiO_3$ (STO) bicrystal as a template for producing a single YBCO [001]-tilt boundary by pulsed laser deposition; the second is an ensemble of YBCO tilt boundaries formed using biaxially aligned polycrystalline substrates. Single grain boundary (SGB) studies give a fundamental approach to GB transport while polycrystalline samples allow for a statistical verification of the developed model, to the extent that the grain misorientation predominantly influences the transport.

The biaxially aligned polycrystalline samples were fabricated using two coated conductor techniques: ion-beam assisted deposition (IBAD)[8] and rolling-assisted biaxially textured substrate (RABiTS).[9] Both architectures were completed with consistently high-J_c, YBCO films produced by the *ex situ* ‘‘barium fluoride’’ process.[10] The RABiTS structure, in this case, uses buffer layers of CeO_2/yttrium stabilized zirconia (YSZ)/CeO_2 on textured Ni (or Ni alloy), which provides an epitaxial template that is both an oxygen diffusion barrier and chemically compatible with YBCO. The IBAD architecture consists of an untextured, metallic alloy substrate coated with ion-beam aligned YSZ and

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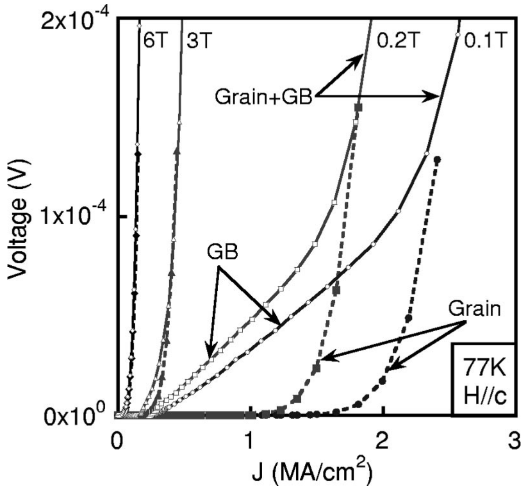

FIG. 1. *V-J* characteristics of a 4.5° SGB. The GB shows NOLD behavior with a transition to nonlinear characteristics as the surrounding grains dominate dissipation. A separate patterning of a grain is also plotted that shows the complete nonlinear *V-J* curve of a grain. Voltage is plotted in place of electric field due to the uncertainty of the width of the GB.

capped with a layer of CeO_2 for compatibility with the *ex situ* BaF_2 process.

Microstructural characterization suggests that the overall distribution of grain boundary tilts in these polycrystalline films are correlated on a local scale.[1] According to electron backscattering maps of the grain misorientation distribution, we define an ''equivalent'' GB angle such that pervasive, long-range connectivity is observed (connecting 70% of the cross section with GB angles less than or equal to θ_{eq}). This enhanced, *local* alignment of grains presumably results from both rolling and ion-beam texturing techniques. Therefore, current transport in coated conductors may be limited by LAGBs of only a few degrees, despite global distributions with full width at half maximum values greater than 10° according to x-ray diffraction.

We previously characterized the *V-J* curves for HAGBs and identified a signature non-ohmic, linear differential (NOLD) behavior.[11] Figure 1 shows both inter-grain and intra-grain dissipation for a 4.5° SGB, with NOLD behavior dominating only up to 4 T. This is a confirmation of a result reported by Diaz *et al.*[12] for a 4° SGB that exhibited clear NOLD behavior at 1.6 T, 77 K. We therefore propose that the transport properties of a wide range of GB angles can be grouped together by a common NOLD signature, which is likely tunneling-dominated behavior. Recall that HAGBs are characterized by a disordered boundary while the low-angle regime shows regions of YBCO that are structurally intact and only periodically punctuated by dislocations. As the GB angle is reduced below 4°, the well-ordered regions between dislocation cores become larger than the coherence length of YBCO,[13] and grain-like dissipation is expected. In this case, we expect that the macroscopic J_c will be fractionally reduced due to the cross sectional loss of supercurrent from dislocations uninhabited by vortices.

To test the strongly coupled model expected at low θ, a nominal 2° GB was measured and showed grain-like *V-J* characteristics and a self-field J_c (77 K) of 4 MA/cm^2. A subsequent 50 μm patterning of the grain and GB confirmed the film homogeneity with nearly identical results. High resolution x-ray diffraction confirmed the expected YBCO misorientations: 1.83° [001] tilt and 0.13° [100] tilt. According to TEM micrographs[14] and the relationship $D = |b|/2\sin(\theta/2)$, the dislocation spacing for a 2° GB is $D \simeq 11$ nm, assuming b is a Burger's vector of 0.4 nm. A maximum strong channel size of 10 nm can be approximated from visual inspection of the dislocation cores from TEM images. Extrapolating, with the assumption of a constant core size, a 4.5° GB would have a strong channel size approximately equal to the 77 K coherence length (3.5 nm). A more elegant approach modeled by Gurevich and Pashitskii[15] predicts nonsuperconducting regions will overlap at 4°, due to excess ion charge. This leads to a view of a 2° GB in which J_c is limited by strongly coupled material punctuated by a periodic array of dislocations that reduce the cross sectional area available for superconducting transport. This is particularly true at low applied fields where numerous dislocations are unoccupied by vortices, due to the large equivalent field (17 T) of the defect density.

If, on the other hand, weak-linked dissipation dominated a 2° GB, simulations using extrapolated values from the HAGB regime, $J_c\,(2\times10^6\ \mathrm{A/cm^2})$ and GB resistivity $(1\times10^{-10}\,\Omega\ \mathrm{cm^2})$, we find that NOLD behavior would clearly be observed in the *V-J* relations. However, if the strong channels of the GB exhibit grain-like, nonlinear *V-J* characteristics, our typical measurement configuration would be insensitive to the small voltage generated at the GB when in series with typical sized grains. This observation is consistent with the results, and proposes the need for a transport-current experiment with voltage probes closer than 10 μm to achieve the sensitivity required to further investigate GBs of a few degrees. The difference in character between the strong and weak-coupled regimes was demonstrated by Polyanskii *et al.*[16] where magneto-optical imaging showed flux penetration into a 5° but not a 3° SGB at 748 Oe (7 K).

Equation (1) phenomenologically quantifies the effect of misorientation angle on J_c for the weakly coupled regime

$$J_c(\theta)=J_c(0)\exp\left(-\frac{\theta}{\alpha}\right);\ \ [\theta\gtrsim4°]. \tag{1}$$

A fit to our SGB data above 4°, using Eq. (1), finds $\alpha=3.2°$ and $J_c(0)=3.2\ \mathrm{MA/cm^2}$, at 77 K, self-field. Published data at 77 K give $\alpha=4.0°$[15] and $\alpha=4.4°$.[5] Recent data at 4.2 K find $\alpha=5.5°$.[17]

Figure 2 combines the SGB data and exponential curve fit from Eq. (1) for comparison with the J_c measurements on both IBAD and RABiTS tapes. These coated conductors give a unique opportunity to verify this low-angle regime with the advantage of statistics and small grain size. In the case of high-purity Ni RABiTS, the grain size typically ranges from 10 to 100 μm, providing sampling of hundreds of thousands of GBs with the applied transport current. This number is at least an order of magnitude larger with IBAD samples, where the grain size is typically less than 1 μm. The present data underscore the important role of biaxial texture by indicating a direct correlation between J_c and the grain-to-grain orientation distribution, as parameterized by the equivalent angle θ_{eq}. This implies that the coated conductors are limited by low-angle GBs and the intra-grain J_c is higher than 3 MA/cm^2. To close the gap between single crystal data and

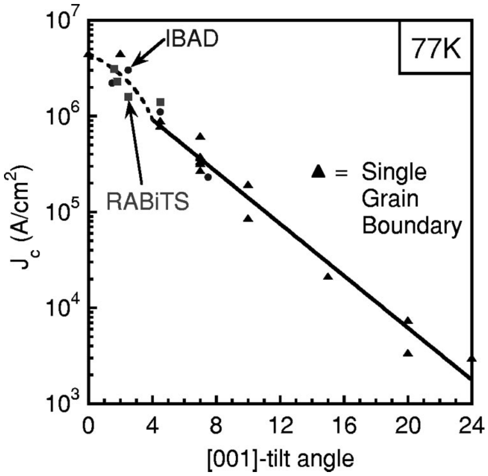

FIG. 2. J_c vs. θ for the SGBs, IBAD and RABiTS samples. An equivalent angle is used in the case of the coated conductors that represents the best estimate of the limiting angle according to electron backscattering diffraction maps. The HAGBs fit an exponential dependence, while the strongly coupled regime below 4° is interpolated with a linear dependence to zero misorientation, 4.5 MA/cm^2, which is represented by both a pulsed laser deposition deposited YBCO/STO and a BaF_2 deposited YBCO/CeO_2/YSZ.

the weak-linked regime at 4°, a linear interpolation is also represented in Fig. 2. This linear dependence on angle is proposed to approximate the expected change in channel size. To reiterate, the data suggest the strongly coupled GBs, found in coated conductors, provide sensitivity to an expected reduction in J_c from disorder along the GB.

Another perspective on tunneling-dominated versus grain-like behavior is illustrated by the results of J_c in magnetic field. Figure 3 shows the 77 K field dependence of J_c for several SGB samples, up to their grain irreversibility field. The field dependence of the GBs are consistent with models[18,19] in which the adjacent flux lattice modifies the equivalent width of, or dephases the order parameter near the GB. This approach yields the field dependence

$$J_c(B)=J_c(0)\left[\frac{B_o}{B}\right]^{1/2}. \tag{2}$$

With an inhomogeneous flux array near the GB, the predicted exponent decreases to 1/4.[18] Fits to a general expression find a range of exponents from 0.63 to 0.15 for 24° to 4.5°, respectively. There is good agreement among all the SGBs for the value of the characteristic field B_o,[11] which implies a constant effective junction area.

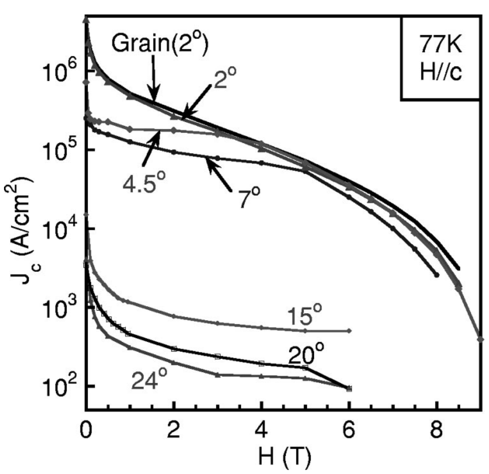

FIG. 3. Magnetic field dependence of several SGB samples up to their irreversibility field. Above 4°, GBs have a reduced J_c and are less sensitive to field than their adjacent grains. At high fields and low θ, J_c is limited by the grains, not the GB.

Of particular interest is the field dependent behavior of the tunneling-dominated 4.5°, and 7° SGBs, which at low field have significantly lower J_c's than their adjacent grains, but like the HAGBs, show less *relative* sensitivity to field than the grains (Fig. 3). A direct result of this field dependence is a range of high fields where the dissipation in the GB is indistinguishable compared to the surrounding grains. This implies that at high fields, the GBs in coated conductors are transparent to transport measurements of loss.

Summarizing, the best coated conductors are characterized by grain-to-grain connectivity of only a few degrees. This can be visualized as grain boundaries perforated by islands of disorder separating channels of strongly coupled superconductor. At small fields, periodic disordered regions will be more numerous than vortices resulting in a reduction in superconducting cross section. However, at high fields, the properties of coated conductors are indistinguishable from single orientation YBCO films. Indeed, the abrupt transition in field dependence, exemplified in Fig. 3, seems to imply that the observed high field J_c is limited by the grain behavior.

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