Critical Currents in [001] Grains and across Their Tilt Boundaries in $YBa_2Cu_3O_7$ Films

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The temperature and magnetic field dependences of the critical-current density of superconducting $YBa_2Cu_3O_7$ films have been determined from transport measurements across individual [001] tilt boundaries and within single [001] grains. The results provide evidence that the basal-plane J_c is limited by flux creep. In contrast, the *I-V* characteristics and the temperature magnetic field dependences of the grain-boundary J_c are consistent with a superconductor–normal-superconductor–type weak-link behavior. These results imply that the transport properties of these materials can be treated within a BCS framework.

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The disappointingly low transport critical-current densities in polycrystalline high-T_c materials have been found to be largely attributable to poor coupling across grain boundaries.[1] Currently, an overall understanding of the mechanisms that limit intragranular and intergranular critical-current densities has not been provided. To elucidate these mechanisms, we report here on measurements of the *I-V* characteristics and of J_c as a function of temperature and magnetic field across [001] tilt boundaries and within the adjacent grains. These dependences provide evidence that the intragrain J_c is controlled by flux creep. For the grain boundaries, the transport properties display a classic superconductor-normal-superconductor (SNS) weak-line behavior. All results are consistent with models based on the BCS theory.

Thin-film bicrystals of $YBa_2Cu_3O_7$, in which two *c*-axis–normal grains were misoriented by an angle θ in the basal plane, were epitaxially grown on $SrTiO_3$ bicrystals, as previously described.[1] The films, which were typically 0.5 μm thick, had a T_c ($R=0$) of $\simeq 88$ K. Critical-current densities were measured on lines, typically 5–10 μm wide and 10–20 μm long, which had been patterned by laser ablation.[2] By patterning a line into each grain and a line across the grain boundary, the intragranular and intergranular critical currents could be directly compared. Critical-current densities were obtained by the division of the maximum zero-voltage current (dc measurement, voltage $<2\ \mu V = 1$ mV/cm) by the cross-sectional area of the lines. As discussed elsewhere,[1] Ohmic heating at the wire bonds did not affect the measurements.

In Fig. 1, the critical-current density is plotted as a function of temperature for two grains. This behavior was displayed by all *c*-axis grains that we measured[3] ($\simeq 15$ samples). Because of the relatively small pinning energies compared to the large superconducting temperature range, it has been suggested[4-7] that the critical currents in high-T_c superconductors should be limited by flux creep, as described by the Anderson-Kim creep model.[8] Yeshurun and co-workers have demonstrated the presence of flux creep in $YBa_2Cu_3O_7$ crystals by measuring their magnetic relaxation rate.[7] However, no evidence based directly on transport measurements has yet been provided for a flux-creep-controlled transport current. Following Tinkham,[6] when the critical current is limited by flux creep, $J_c(B,t)$ is given by

$$J_c(B,t) = J_c(B,0)[1-\alpha(B)t-\beta t^2] \tag{1}$$

for $t = T/T_c \ll 1$, where B is the magnetic flux density. The coefficient β in Eq. (1) comes from the temperature dependence of the free-energy difference between pinned and unpinned flux quanta, expanded in the form $U(B,t) \simeq U(B,0)(1-\beta t^2)$. The coefficient $\alpha(B)$ is given by

$$\alpha(B) = \frac{kT_c}{U(B,0)} \ln\left(\frac{aB\Omega}{E_{\min}}\right), \tag{2}$$

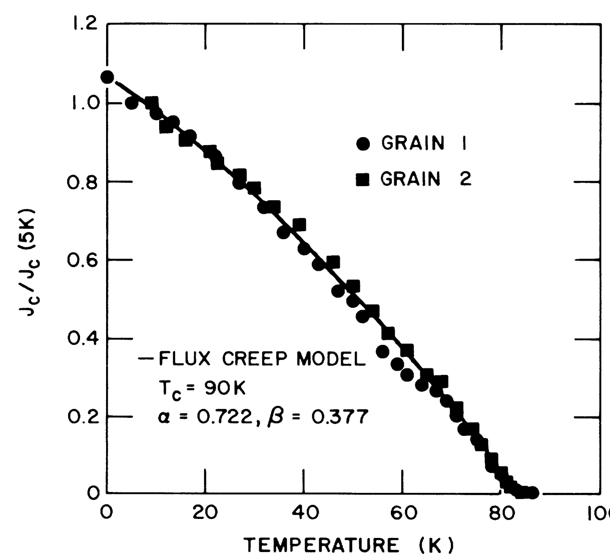

FIG. 1. Normalized critical-current densities vs temperature for two *c*-axis grains. The solid line is a fit by the flux creep model. $J_c(0)$ equals roughly 6×10^6 A/cm^2 for both grains.

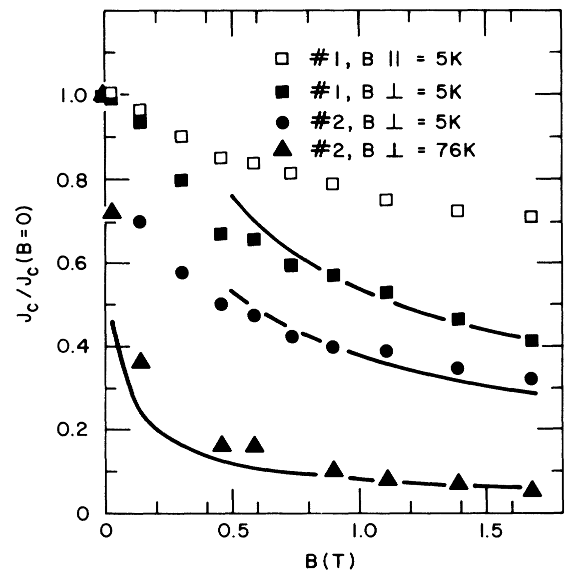

FIG. 2. Normalized basal-plane critical-current densities vs B for two samples. The magnetic fields are oriented to the film plane as indicated, but are always perpendicular to the transport current. $J_c(0)$ equals 1.3×10^6 and 4×10^6 A/cm^2 for samples No. 1 and 2, respectively.

where a is the average hopping distance of the flux quanta, Ω is the attempt frequency for escape, and $E_{\min}$ is the electric field criteria that defines J_c. As illustrated in Fig. 1, Eq. (1) fits $J_c(0,t)$ well for $\alpha=0.72$ and $\beta=0.38$, except near T_c. Approximating Ω by typical phonon frequencies of 10^{10} Hz,[7] B by the average self-field of the transport current, and a by the average flux line spacing,[4] Eq. (2) yields a pinning energy $U(0,0)$ of 70 meV. This value compares favorably with the value of 150 meV derived from relaxation data.[7]

The flux creep model also accounts for the behavior of J_c in magnetic fields. If an applied magnetic field results in a flux lattice, $J_c(B,t)$ is given by

$$J_c(B,t)=\frac{N_pU(B,0)}{1.07(\Phi_0B)^{1/2}}[1-\alpha(B)t-\beta t^2]\,, \qquad (3)$$

as pointed out by Dew-Hughes.[4] Here N_p is the density of the pinning sites and Φ_0 is the flux quantum. The dependence of $J_c(B)$ is shown in Fig. 2 for fields applied both perpendicular and parallel to the plane of the film. The critical-current density follows a $B^{-1/2}$ behavior, which is predicted by Eq. (3), if small effects due to the magnetic field dependence of the logarithmic term in α and due to $U(B,0)$ are neglected.[9] Deviations (from $J_c\propto B^{-1/2}$) at small fields can be attributed to the self-field of J_c, which will decrease with J_c for increasing temperature, as observed.

The slope of the normalized critical-current density with respect to temperature increases at low temperatures with increasing magnetic field, as illustrated in Fig. 3. This behavior is consistent with the creep model as

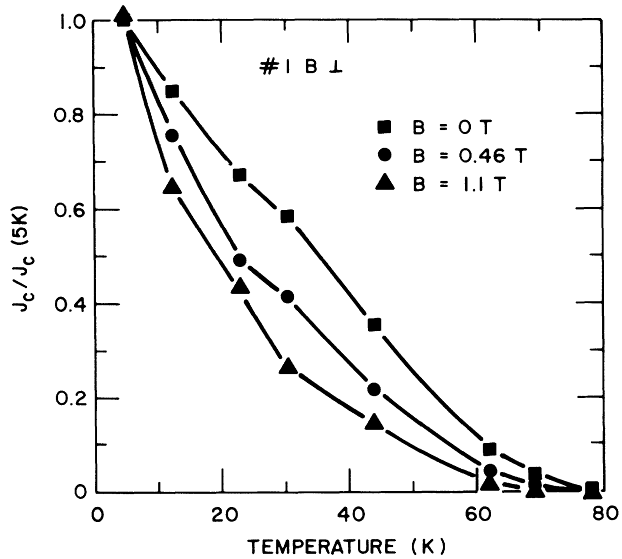

FIG. 3. Normalized critical-current density vs temperature for sample No. 1 (compare with Fig. 2) in magnetic fields perpendicular to the film. This particular sample showed a pronounced tail in the $J_c(T)$ characteristic for $T>65$ K.

seen by differentiating Eq. (3):

$$\frac{\partial[J_c(B,t)/J_c(B,0)]}{\partial t}=\alpha(B)\,,$$

for $t\ll1$. However, the full temperature dependence for $B\gg0$ is not easily accounted for. The I-V behavior of the grains is characterized by a gradual voltage onset and an increasing $dV/dI(V)$ at all voltages; this behavior is also consistent with that expected for a creep-limited sample. It should be noted that the agreement between the intragranular transport characteristics and the Anderson-Kim model implies that these characteris-

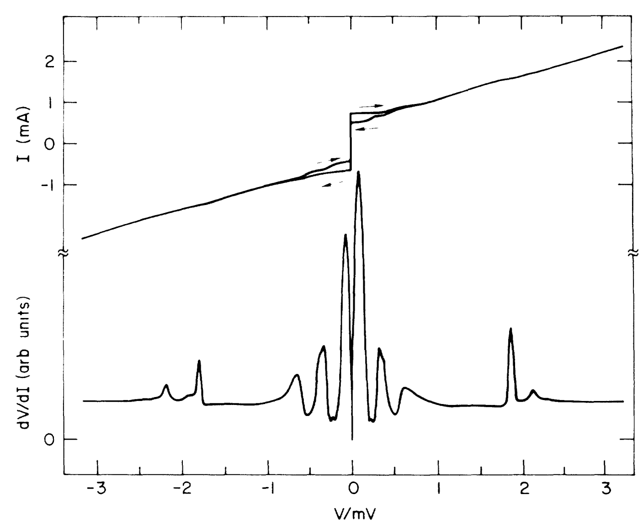

FIG. 4. $I(V)$ and $dV/dI(V)$ characteristic at 5 K for a 37° grain boundary.

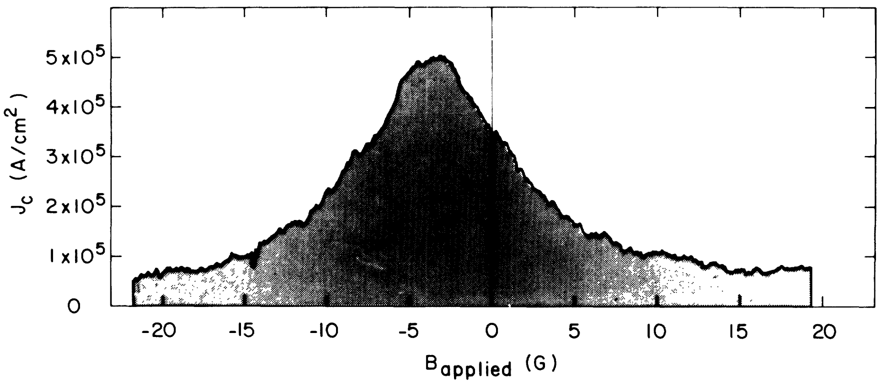

FIG. 5. Critical-current density vs applied magnetic field (perpendicular to the film) at 5 K for a 5° grain boundary.

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The $I(V)$ characteristics of grain boundaries with misorientation angles $\gtrsim 5°$ (Fig. 4) differ from those of the grains. Once J_c is exceeded, the voltage jumps to a finite value, often displaying hysteresis. For higher currents, the normal-state resistance is roughly constant. This behavior, which is found over a wide temperature range ($t \lesssim 0.8$), suggests that the intergrain J_c is not creep limited. In fact, all the features observed in the $I(V)$ characteristics, even the bumps at low voltages seen in Fig. 4, are well known for conventional weak links.[10]

The variation of J_c across a grain boundary as a function of applied magnetic field is shown in Fig. 5 for a misorientation angle of 5°. The asymmetry about $B=0$ was observed only after a magnetic field had been applied, and is, thus, attributed to trapped flux. It should be noted, that even for this relatively low-angle boundary, J_c(20 G) is reduced by almost an order of magnitude from J_c(0 G). This extreme sensitivity of J_c to small magnetic fields, which is characteristic of all grain boundaries, further demonstrates their weak-link nature. Interestingly, the critical current becomes less sensitive to the magnetic field as the field is increased (Fig. 5).

The temperature dependence of J_c across boundaries with misorientation angles $\gtrsim 5°$, follows a universal curve (Fig. 6).[3] This behavior differs from the creep-limited $J_c(T)$ of the grains. The temperature dependence of the normalized J_c can be accurately fitted (Fig. 6) with the Ambegaokar-Baratoff equation for the critical-current density of a Josephson junction,[11]

$$J_c(T) = \frac{\pi\Delta(T)}{2eR_{NN}} \tanh\left[\frac{\Delta(T)}{2kT_c}\right], \tag{4}$$

where $\Delta(T)$ is the gap, and R_{NN} is the specific tunneling resistance, which is assumed to be temperature independent. Good agreement between Eq. (4) and the experimental data is obtained only if a gap $\Delta(0)$ of about 5 meV is used. It is conceivable that the grain boundaries are superconductor-insulator-superconductor-type Josephson junctions, to which Eq. (4) is evidently applicable; however, no microscopic evidence for an insulating layer at the boundary was found.[1] If the boundaries are superconductor-insulator-superconductor-type junctions, Δ reflects the value of the gap in the $YBa_2Cu_3O_7$ near the boundary, which might be lower than the gap in the rest of the film. On the other hand, Eq. (4) is also valid for dirty SNS junctions, provided that significant changes in the phase occur only near or in the normal layer. In this case, the observed gap represents the reduced order parameter inside the normal layer. The current density $J_c(0)$ that is predicted by Eq. (4) is an order of magnitude higher than the measured $J_c(0)$. Although the grain boundaries are large Josephson junctions, this effect would not be caused by the self-field effect of the Josephson current, which is small for the

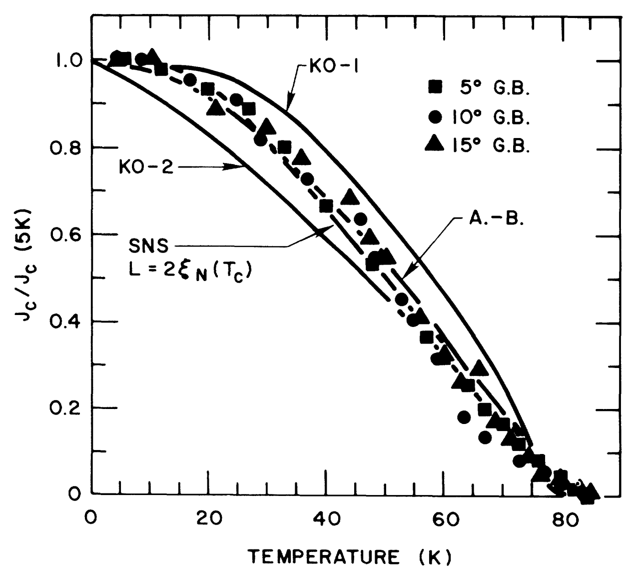

FIG. 6. Normalized critical-current density vs temperature for three grain boundaries. KO-1 and KO-2 are the dependences predicted by Kulik and Omel'yanchuk for weak lines in the dirty and in the clean limits, respectively. The curves labeled AB and SNS refer to the theories of Ambegaokar-Baratoff and Likharev, respectively.

grain-boundary geometry.[12] In fact, the low $J_c(0)$ might indicate strongly coupled superconductors. It is also worth noting that the I_cR_{NN} products of all boundaries ranged between 0.3 and 3.4 meV with an average of 1.5 meV.

Besides the Ambegaokar-Baratoff theory, other models which predict $J_c(T)$ for weak links may be applicable. For example, Kulik and Omel'yanchuk have calculated $J_c(T)$ for short weak links, such that $L/\xi \rightarrow 0$, where L is the length of the weak link.[13] While their predicted temperature dependence is in reasonable agreement with the experimental results (Fig. 6), it is questionable if $L/\xi \rightarrow 0$ is a good approximation, since the thickness of the grain boundary, $L \simeq 1-2$ lattice constants, and the coherence length ξ are comparable. Likharev[14] has calculated the $J_c(T)$ dependence of SNS junctions for arbitrary $L/\xi_N(T_c)$ ratios, where $\xi_N(T_c)$ is the coherence length of the normal layer, which is estimated[15] to be roughly 5 Å for a grain boundary. The temperature dependence calculated by Likharev agrees well with the experimental data (Fig. 6) for $L/\xi_N(T_c) = 2$. It should be noted that this agreement is achieved with a BCS gap of $\Delta(0) = 1.76kT_c$. Interestingly, a T_c has to be assumed which is 10% lower than that of the grains of the same film, which suggests that the order parameter of the grains is depressed near the boundary. Near T_c, the bulk coherence length becomes large, so that this depression will be less severe. This reasoning could account for the tail ($T > 80$ K) in J_c.[16] Again, the predicted $J_c(0)$ according to these theories is an order of magnitude higher than the measured values. It should be noted that the transport characteristics of the grain boundaries are successfully described by standard theories for weak links and are, thus, consistent with the BCS theory.

In conclusion, the intragranular (basal plane) critical-current density is found to be flux-creep limited. Although the critical-current densities of epitaxial films are already high ($J_c > 10^6$ A/cm^2 at 77 K), this conclusion suggests that it might be possible to increase J_c further by introducing defects with large pinning energies. In contrast, the critical-current density across a grain boundary is limited by poor coupling across this distorted interface and not by the pinning properties of the boundary. This implies that doping the grain boundary and thereby creating additional electronic states or enhancing the proximity effect might increase J_c. Finally, we have shown that the observed transport characteristics of the grains and their boundaries are entirely consistent with the conventional BCS theory of superconductivity.

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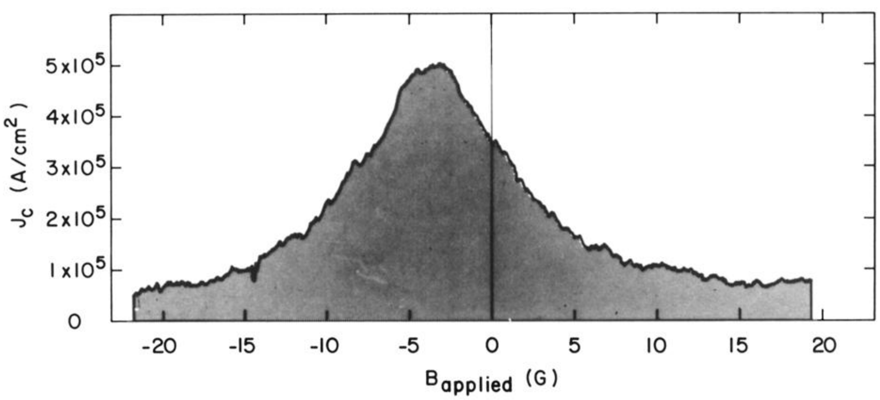

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