Scaling behavior of YBa$_2$Cu$_3$O$_{7-\delta}$ thin-film weak links

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(Received 21 June 1990; accepted for publication 6 July 1990)

The superconductive weak link properties of microbridges formed in c-axis normal YBa$_2$Cu$_3$O$_{7-\delta}$ polycrystalline thin films containing a variable amount of large angle tilt boundaries have been studied. In the low critical current density limit these weak links have current-voltage ($I$-$V$) characteristics that are accurately modeled by the resistively shunted junction model. The $I$-$V$'s are found to accurately follow a simple scaling law with the product of the critical current and weak link resistance $R_s$ varying linearly with the weak link conductance.

Due to the strongly anisotropic nature of the cuprate high-temperature superconductors (HTS) it appears necessary that to achieve the highest critical current density $J_c$, the predominant direction of current flow must be constrained to that defined by the copper oxide planes. In addition it appears that tilt boundaries, formed by the junction of two rotated c-axis aligned grains, will also result in a major, although not necessarily fatal, reduction in the maximum $J_c$ that can be sustained across the boundary. Despite extensive work investigating the properties of individual tilt boundaries in YBa$_2$Cu$_3$O$_{7-\delta}$ thin-film bicrystals, the microscopic nature of the degrading effect of these tilt boundaries has not yet been established, nor has a clear determination been made as to whether this degradation is fundamental to YBa$_2$Cu$_3$O$_{7-\delta}$ grain boundaries.

Here we report on the results of a series of measurements of the superconductive properties of thin-film microstructures that have been formed from c-axis normal oriented, polycrystalline YBa$_2$Cu$_3$O$_{7-\delta}$ thin films. These microbridges are sufficiently small, and the mean grain size of the film sufficiently large, that a single grain boundary weak link in the microbridge usually determines its superconductive transport properties. We find that these weak links, if of low critical current density, have the current-voltage ($I$-$V$) characteristics of an essentially ideal resistively shunted junction (RSJ) Josephson element, while weak links with higher $J_c$ have characteristics attributable to one-dimensional flux creep and flux flow. We have used these weak links in experiments in which the oxygen content, and hence presumably the hole carrier content, of the film is reversibly varied by short low-temperature anneals. We find that the $I$-$V$'s of the weak links before and after anneal are essentially identical if the voltage and current are properly scaled. This and the closely related regular variation of $I$-$R_s$ of the weak links, where $I$ is the critical current and $R_s$ the normal resistance, with the weak link conductance per unit area $\sigma$, strongly suggest that the suppressed superconductive transport properties of the grain boundaries are quite systematic and perhaps fundamental.

The films employed in this study were formed by laser ablation on oriented single-crystal MgO (100) substrates using now routine process procedures that are described elsewhere. Depending upon the substrate preparation and process parameters we find that films can be produced that are fully oriented with the c axis normal to the (100) MgO substrate, but that contain a variable component of large angle grain boundaries due to the YBa$_2$Cu$_3$O$_{7-\delta}$ grains assuming several alternative rotations about the c axis. Through x-ray pole figure analysis we find that the preferred orientation is with the a and b axes aligned with the MgO cubic axes in the plane of the substrate. But in addition there are grains rotated 45° (about the c axis) from the preferred orientation. The percentage of these 45° grains can vary from greater than 20% to 0% depending upon the details of the growth process. The pole figure data have been confirmed by scanning transmission electron microscopy (STEM) plan view and cross-section studies of the YBa$_2$Cu$_3$O$_{7-\delta}$ films.

[FIG. 1. (a) $I$-$V$ characteristic of a low critical current density weak link in a ~2-µm-long, ~2-µm-wide, 300-nm-thick YBa$_2$Cu$_3$O$_{7-\delta}$ microbridge taken at 4.2 K. The dotted line is a resistively shunted junction model fit to the data, where a sinusoidal supercurrent phase relation, a zero shunt capacitance, and a linear quasiparticle resistance are assumed. (b) $I$-$V$ characteristic of a 10-µm-long, 0.8-µm-wide microbridge which shows the presence of three weak links in series. The dotted line is a series RSJ fit.]
Films approximately 300 nm thick have been patterned using conventional photolithography and inert ion milling to form multiple, rectangular microbridges on a given substrate, where the width of the microbridge ranges from 0.8 to 3 µm and the length from 2 to 10 µm. In general we find that the superconductive properties of the narrower microbridges are dominated by one or more weak links. On a given film the critical current density of these weak links can vary by as much as two orders of magnitude. Examples of the $I$-$V$'s that are obtained with these weak links are shown in Figs. 2 and 3. If the critical current density is sufficiently low, $\sim 10^4$ A/cm$^2$, that the effective penetration depth of the weak link is greater than the width of the microbridge, then we find that the $I$-$V$'s are closely modeled by the resistively shunted junction (RSJ) model. This is illustrated in Fig. 1(a). In Fig. 1(b) we show the $I$-$V$ that was obtained from a comparatively long, 10 µm, 0.8-µm-wide microbridge which clearly shows the presence of three separate weak links, each with a different critical current and normal-state resistance. Again a simple series RSJ model is a very good approximation to the observed $I$-$V$ characteristic.

Changes occur in the $I$-$V$ characteristics as $J_c$ of the weak link becomes progressively larger. First we see the situation where the voltage variation quickly takes on a linear behavior for $I > I_c$, but shows an “excess current.” The most plausible explanation for this is similar to that provided by Waldram et al., for the $I$-$V$ response of wide, overdamped Josephson junctions. For weak links with still higher values of $J_c$ (\(\sim 10^6\) A/cm$^2$ at 4.2 K) the voltage onset across the weak link occurs at a well-defined critical current but in a nonlinear manner as illustrated in Fig. 2(b), which is indicative of the onset of one-dimensional flux creep along the weak link.

An experiment was undertaken in which oxygen was reversibly removed from the patterned films after $I$-$V$ characterization. This oxygen removal is essentially complete after a 120 s low-temperature, 500 °C, inert gas annealing of the films, after annealing, and after a 1 min, 500 °C reanneal in oxygen.

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FIG. 2. Scaled $I$-$V$ characteristics of two $YBa_2Cu_3O_7$ weak links taken at 4.2 K, before a 120 s low-temperature, 500 °C, inert gas annealing of the films, after annealing, and after a 1 min, 500 °C reanneal in oxygen.

FIG. 3. Variation of 4.2 K weak link $J_cR_n$ with conductance per unit area for a number of weak links found in microbridges formed from polycrystalline c-axis oriented $YBa_2Cu_3O_7$ films laser ablated onto MgO. The least-squares fits to the data including the point indicated by $A$ yields a slope of 0.85, excluding that point, which might be attributable to two nearly identical weak links in series, yields 0.97.

 characteristics of a number of individual weak links were first measured. By heating the films for approximately 10 s at 500 °C in an inert atmosphere, a sufficient amount of oxygen is removed from the film to increase the bulk film room-temperature resistivity, which is not dominated by the resistance of the weak links, by approximately a factor of 2. Comparing the accompanying shift in c-axis lattice constant with published values we estimated $\delta$ to have been changed from 0 to 0.1 by the anneal. This oxygen removal is believed to occur predominantly at the copper oxide chain sites which lie along the $b$ axis in the orthorhombic phase of $YBa_2Cu_3O_7-b$. The simplest view of this oxygen removal process would be that it allows us to significantly change the carrier density in the film without otherwise materially altering the thin-film microstructure.

In general this inert gas anneal step is sufficient to reduce the effective $T_c$ for the detectable onset of a supercurrent of a particular weak link by nearly a factor of 2. There is an accompanying reduction in $J_c$ of the weak links and a rise in $R_n$. This oxygen removal is essentially completely reversible—a short, 1 min anneal at 500 °C in flowing oxygen is usually sufficient to return the effective $T_c$, $J_c$, and $R_n$ to very near their original values.

We have found a remarkable scaling behavior in the weak link $I$-$V$ characteristics. In particular if the weak link voltage is scaled by $I_c^{1/2}$ and the current by $I_c^{-1}$, the low $T_c$ 4.2 K, $I$-$V$'s of any individual weak link before and after oxygen removal are essentially identical. Two examples of this scaling behavior are shown in Fig. 2. We find that this scaling applies to weak links whose $I$-$V$ character ranges from that indicative of simple RSJ-like behavior to that of one-dimensional flux creep and flux flow. Thus we find that the effective dynamic resistance of the weak links is directly, and quite precisely, proportional to $I_c^{1/2}$.

This $I_c \sim R_n^{-1/2}$ scaling is also found to apply, albeit less precisely, to the $I_c$'s of a number of different weak links fabricated in a group of films all grown in a similar manner. This is shown in Fig. 3 where $I_cR_n = J_cR_n$ is plotted versus the conductance per unit area, $\sigma_n = 1 / \rho_n = 1 / R_nA$, for a number of such weak links. Here the variation in $J_c$ is limited to values $\leq 10^4$ A/cm$^2$, since only then do the $\sim 1$-µm-wide weak link $I$-$V$ characteristics generally exhibit a linear behavior from which an effective normal
resistance can be extracted. While there is considerable scatter in the data of Fig. 3, there is clearly a general increase in \( I_eR_e \) with increasing \( \sigma_n \), with a linear least-squares fit to the data yielding \( I_eR_e \propto \sigma_n^{0.45} \).

The microscopic nature of these weak links is uncertain. The magnetic field dependence clearly shows that the weak links are quite inhomogeneous in the directions perpendicular to the current flow, despite showing good RSJ-like behavior. As reported by others,\(^2\) we find that \( R_e \) of the weak links, when it can be clearly determined, is essentially temperature independent. Also, the low-temperature variation of \( I_e \) is quite similar to the nearly linear variation of the bulk film \( I_e \) with decreasing \( T \) found in measurements on quasi-single-crystal thin films. Accordingly, the grain boundary contact can then either be\(^7\) (a) an inhomogeneous, thin superconductor-normal metal-superconductor SNS contact where the thickness of the normal region \( d_n \ll \xi, \) where \( \xi \sim 0.18hV/\hbar kT \) is the normal metal coherence length, (b) an inhomogeneous insulator tunnel (SIS) junction for which the quasiparticle resistance is linear, with no indication of a gap in the density of quasiparticle states, or (c) an inhomogeneous weak S-S'-S superconducting contact whose resistance in the S' region is dominated by very strong, temperature-independent impurity scattering. One particularly simple version of this last case would be a parallel array of ideal ballistic point contacts, denoted as ScS, extending through an otherwise insulating grain boundary.

If the weak link is an SNS junction,\(^9\) the measured weak link resistance would then seem to require the resistivity of the normal region to be of the order of 0.1 \( \Omega \) cm, which implies the barrier is at best a semiconductor and that the contact is basically a tunnel junction.

Both an ScS contact\(^9,10\) and an SIS tunnel junction\(^9\) are expected to have a \( T = 0 \) value of \( I_eR_e \sim 2\Delta/e \), where \( \Delta \) is the value of the pair potential or energy gap of the superconductor adjacent to the contact or tunnel barrier. But \( \Delta \) for \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) is expected to be and perhaps has been measured\(^11\) to be \( \gtrsim 15 \) mV. The very low values of \( I_eR_e \) that have been observed in the \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) weak links could be attributed to the effect of some low resistance shunting of the tunneling barrier, or to a greatly depressed value of \( \Delta \) at the edge of the weak link, perhaps due to an extreme carrier deficiency in the film at the grain boundary, or to the presence of a very strong pair breaking process at the grain boundary. The accurate scaling of \( I_eR_e \) with \( \sigma_n \), as well as much of the HTS weak link data that can be found in the literature, suggests that whatever the mechanism, the reduction in \( I_eR_e \) to \( \ll 0.1 \) of its expected value is a very systematic and regular effect and thus is not due to some uncontrolled resistive shunting of the weak link.

We note that if we wish to treat the weak link as an otherwise ideal Josephson-like contact, but with a greatly reduced energy gap \( \Delta \) at the electrode-weak link interface, then for the SIS (or S-semi-S)-case\(^3\) we must have that 
\[
\Delta \propto \sigma_n \sim T^2 N(0)V_F \rho_F^2, 
\]
where \( T^2 \) is the tunneling transfer matrix element and \( N(0) \) is the local density of states. If, alternatively, the weak link is an ScS contact of some type then the scaling indicates that 
\[
\Delta \propto \sigma_n \sim \chi N(0)V_F \rho_F^2, 
\]
where \( \chi \) is the local Fermi velocity. Here, based on the stem studies, we have modeled the weak link as simply causing a very localized reduction in the maximum possible (ballistic) current density, and thus \( \chi \) is the mean probability for an incident charge to be transported across the grain boundary without backscattering. Since the removal of oxygen from the CuO chain sites is expected to reduce \( N(0) \), an expectation which is supported by the resulting increase in the bulk film resistivity, an interpretation of the observed \( I-V \) scaling as indicating a linear variation of \( \Delta \) with \( N(0)V_F \) is certainly consistent with the data for either case. However, this would not appear to be compatible with a BCS-like model of high-temperature superconductivity.

If, instead, we wish to invoke a very strong pair breaking process at the weak link as the cause of the local reduction in \( \Delta \), then \( \sigma_n \) must be dominated by pair breaking so that 
\[
\Delta \propto \sigma_n \sim \Gamma, 
\]
where \( \Gamma \) is the pair breaking scattering rate. Then the effect of the oxygen removal would be interpreted as chiefly increasing \( \Gamma \) at the grain boundary and not affecting \( N(0) \) significantly. In this context, it is interesting to note that if we estimate the ideal ballistic conductance per unit area of \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) we find that 
\[
\sigma_{\text{ideal}} \approx N(0)\rho_F^2/3 \times 4 \times 10^{10} \text{S/cm}^2, 
\]
where we have assumed\(^12\) \( N(0) \approx 3.5 \times 10^{27} \text{eV} \text{cm}^{-3} \) and \( \rho_F \approx 2 \times 10^{-7} \text{cm/s} \). From our measured variation of \( I_eR_e \) with \( \sigma_n \), this implies that a very short "weak link" in a fully stoichiometric \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) microbridge with a uniform and ideal ballistic conductance, i.e., no impurity scattering would have \( I_eR_e \approx 100 \text{ mV} \), which is about a factor of 3 higher than would be expected from a BCS superconductor with \( T_c = 90 \) K. This would then suggest that most of the scattering at the weak link results in pair breaking.

This research was supported by the Office of Naval Research (N00014-89-J-1692) and by the Defense Advanced Research Projects Agency (N00014-88-K-0374). Additional support was received from the National Science Foundation through use of the facilities of the National Nanofabrication Facility and the Cornell Materials Science Center.