High field behavior of artificially engineered boundaries in melt-processed $YBa_2Cu_3O_{7-\delta}$

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Artificial bulk "zero-angle" boundaries parallel to the c axis have been engineered between large melt-processed $YBa_2Cu_3O_{7-\delta}$ (YBCO) grains and observed to carry a transport supercurrent at fields up to at least 5 T at 77 K. The temperature and angular dependencies of the boundary resistance have exactly the same form as those of the grains, which is evidence that the grains are intimately coupled. The limiting mechanism for current transfer across these boundaries is, therefore, not a simple weak link or Josephson effect. This joining technique is extremely promising for production of macroscopic engineering artifacts. © *1998 American Institute of Physics.* [S0003-6951(98)00327-1]

The behavior of grain boundaries in high temperature superconductors is of considerable fundamental interest and technological importance. The absence of such boundaries is associated generally with enhanced current carrying ability and it has been demonstrated that melt-processed YBCO (MP-YBCO) can trap fields of 2–3 T at 77 K.[1] Fields of this magnitude exceed those generated by rare-earth permanent magnets and are applicable to a range of possible devices. The maximum field attainable in a superconductor is determined by the product of the critical current and the radius of the current loop (the sample size in the best case). MP-YBCO typically exhibits critical current densities J_c of order 10^5 A/cm^2 at 77 K (in small samples ≈ 50 mm^3) which generate the maximum field values attainable in the material. J_c often decreases with radial distance from the center of the sample, however, due to microstructural inhomogeneities.[2] Further, the diffusion rates and associated long growth times hinder production of samples of diameter greater than 5–7 cm.[3] As a result, the development of a suitable joining technique would represent significant technological progress. Very few attempts have been made to achieve this. Salama and Selvamanickam[4] have joined samples parallel to the c axis and report a J_c of 2000 A cm^{-2} at 77 K in 1 T. Shi[5] has also joined YBCO samples and found qualitative evidence of strong intergrain coupling. Here we report the results of a different joining technology and show that the field dependence of the irreversibility line (IL) of the artificially produced grain boundaries replicates that of the grains in fields of up to 7 T applied parallel to both ab and c directions. Implications for the current limiting mechanism are discussed.

Samples of seeded MP-YBCO were prepared following Ref. 6. Several pieces were cut from a single large grain monolith. The ab planes were identified using the orientation of the platelet boundaries.[6] The c axes of samples in which no macroscopic cracks could be detected by optical microscopy were aligned to form a "zero-angle" boundary normal to the ab planes, as shown in the inset to Fig. 1. This sample was then processed at 920–980 °C in air under a uniaxial pressure of 0.5 MPa to induce recrystallization at the grain–grain interface. Because this treatment modifies the oxygen content of the material substantially, the sample was reannealed at 420 °C for 100 h in oxygen to restore it to close to optimal doping. The detailed joining process is described elsewhere.[7] Narrow bars were cut from the joined sample with the artificial grain boundary (GB) lying across the specimen center. These bars were mechanically polished to produce samples of approximately 4 mm×0.6 mm×0.4 mm. Contacts were deposited by thermal evaporation of silver followed by application of silver epoxy, annealed at 420 °C in oxygen for 5 min. The resistance of the contact pairs was less than 1 Ω. Contacts were applied on either side of the GB, as well as within one or both grains (see Fig. 1). This enabled the resistance across the boundary and within the grain (R_{ab}) to be measured simultaneously. An ac measurement technique at 72 Hz using low noise transformers and

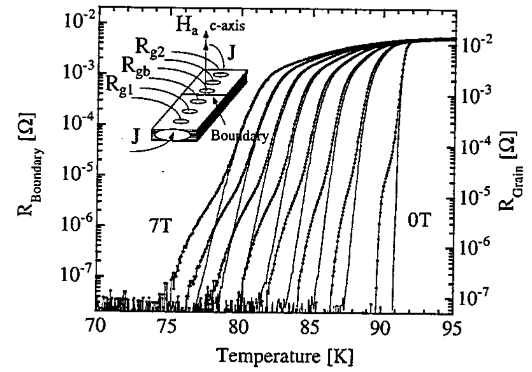

FIG. 1. Resistance as a function of temperature with a current of 20 mA for fields of 0, 1, 2, 3, 4, 5, 6 and 7 T applied parallel to the c axis for the grain (solid lines) and the boundary (marked by points). The inset shows the contact configuration.

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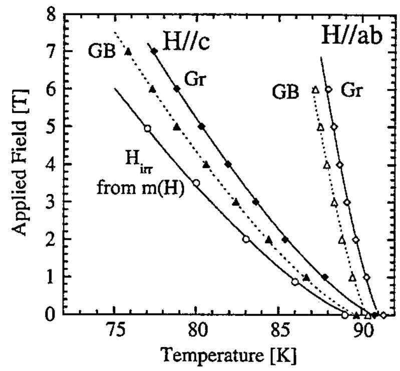

FIG. 2. Irreversibility lines extracted from the data in Fig. 1 (closed diamonds, grain and closed triangles, boundary) and similar data for the field applied parallel to the ab planes (open diamonds, grain and open triangles, boundary). The curves fits for the boundary (dashed) and grain (solid) are described in the text. The irreversibility line for a grain (cut from one end of the same sample), determined from magnetic moment measurements with $H\|c$ is indicated by the open circles. The line in this case only is a guide.

lock-in amplifiers was employed. Magnetic fields of up to 7 T were applied relative to the c axis or ab planes, respectively. Three samples cut from two different joined specimens were characterized to confirm reproducibility of the results. Although the irreversibility field of one of the samples was about 0.8 of the other two, all of the general characteristics were the same. Hence, the following discussion concentrates on the behavior of the best sample. Isothermal magnetization measurements were also made with a quantum design superconducting quantum interference device magnetometer using a 3 cm scan length to minimize the effects of field inhomogeneity.

Figure 1 shows the field-dependent broadening of the resistive transition measured across an artificial GB together with that of an adjacent grain for $H_a\|c$. The applied current was 20 mA ($J\approx 10$ A cm^{-2}). The normal state resistance of the grain is metallic with a small residual resistance ratio $R_{300\,K}/R_{0\,K}$, obtaining $R_{0\,K}$ by extrapolation of the normal state resistance, indicating the high quality of the parent material.[8] The T_c onset value of $\approx$91.5 K is comparable with the best values obtained in MP-YBCO. The transition for the grain is very sharp and broadens with increasing magnetic field in a similar manner to that observed in YBCO single crystals.[9] The behavior of the GB is shown on the same plot. This is characterized by an initial rapid decrease in resistance, which is associated with the adjoining grains, followed by a distinct "foot" in the transition which characterizes the transition of the GB. Both the initial decrease and the foot broaden in a similar manner to the grain with increasing applied field. Figure 1 shows that apart from the GB having a slightly lower T_c than the grain, the field-dependent behavior of the two are very similar in all other respects. Qualitatively identical results were obtained when the magnetic field was aligned with the ab plane.

The data in Fig. 1 can be used to define a "resistive irreversibility temperature" above which the grain and the GB can no longer sustain a supercurrent. This is extracted using a criterion of $2\times 10^{-7}\ \Omega$ and presented in Fig. 2 for both orientations of applied magnetic field. We first concentrate on the behavior of the grain. The weak upward curvature is very similar to that of the melting transition observed in single crystals. The "irreversibility" and "melting" fields have been found to be coincident in twinned YBCO crystals.[10] The melting field in YBCO single crystals for $H\|c$ is expected to occur (for optimally doped material) at 9 T at 77 K. It obeys a power-law relation given by $H_m=H_0(1-T/T_c)^n$ (Ref. 11) with $\mu_0H_0=87$ T, $T_c=92$ K and $n=1.24$ for $H\|c$. The IL here lies below this locus. A good fit of the above equation to the data in Fig. 2 can be obtained, for H_{irr} instead of H_m, with $n=1.33$ and $\mu_0H_0=89$ T and $T_c=90.8$ K for $H\|c$ and $\mu_0H_0=500$ T and $T_c=91.1$ K for $H\|ab$. This suggests that the lines have a very similar form to that reported in Ref. 11 but are suppressed to lower temperatures because of the slightly lower oxygen content of the grains. The anisotropy can be estimated from the ratio between the lines for $\mu_0H_{irr}\|ab$ and $\|c$, respectively, using anisotropic scaling theory which has been shown to be valid in YBCO crystals.[9] Using the formula from Ref. 12, $H_{irr}(\theta)=H_{irr}(\theta=0)/(\cos^2\theta+\gamma^2\sin^2\theta)^{1/2}$, where γ is the mass anisotropy and θ is the angle between the applied field and the ab planes, we obtain a value of $\gamma=5.6$ for our sample. This is within the range expected ($\gamma=5-7$) for near optimally doped single crystals.[12]

Next we turn to the behavior of the GB. This is almost identical to that of the grain but displaced to lower temperatures by less than 2 K. It can be fitted to the same expression as for the grain, with the same value of n and only slightly different values of $\mu_0H_0=83$ T, $T_c=89.8$ K for $H\|c$ and $\mu_0H_0=480$ T and $T_c=90.4$ K for $H\|ab$. This suggests that the same mechanism determines the current carrying behavior of the boundary and the grains, even at high fields and is the central result of this letter. The slightly different values of T_c for the boundary and grain may result from the different activation energies for oxygen diffusion in the boundary relative to the grain.[13] However, effects of cation contamination, strain or dislocations and stacking faults at the boundary cannot be precluded as other possible explanations.

The field-dependent "resistive irreversibility temperature," as defined above, represents an upper limit to the true IL where the linear resistance vanishes and a critical current appears. The resistances, both of the grain and the GB, are still Ohmic and display arrhenius behavior at the point where the signal vanishes below our resolution and where we define the irreversibility temperature. If we employ a less stringent (i.e., larger) voltage criterion, the irreversibility temperature is shifted to higher temperatures and vice versa. What is really measured, therefore, is a line of constant but small resistance, which lies close to but *above* the true IL. Magnetization measurements, on the other hand, enable a lower limit for the IL to be obtained. Magnetization loops at fixed temperature allow a temperature dependent "magnetic irreversibility field" to be defined where the critical current drops below an appropriately chosen critical current criterion. It is, therefore, a line of small but constant critical current which lies close to but *below* the true IL. This magnetically determined irreversibility line for a grain cut from one end of the same sample, as in Fig. 1, is also shown in Fig. 2. It is obtained using a criterion of 25 A/cm^2 which is comparable with the typical currents used in the transport measurements. Although the current densities in the two measurements are similar, the effective electric field in the magnetic

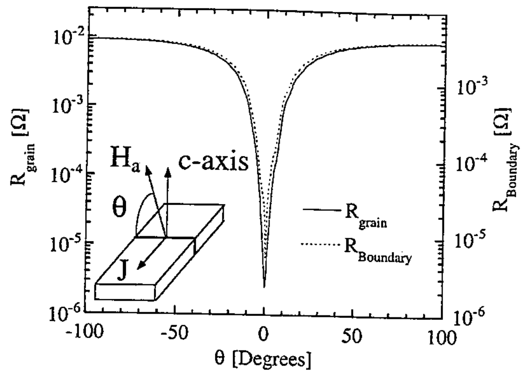

FIG. 3. Angular dependence of the resistance of the grain (solid lines) and boundary (dashed lines) measured at 89.3 K and 3 T when the field is rotated through alignment with the *ab* planes (as shown by the schematic in the inset).

measurement is much smaller. Despite this, the two lines differ only by a factor of 0.3 at 77 K. This gives us confidence that the transport measurements for the grain, and therefore, also for the GB, are a reasonable estimate of the temperature at which a critical current appears. The critical current of the grain was also determined from the same data using the Bean model and yields a value of 2.2×10^4 A/cm^2 at 77 K and 1.8 T, which is a further indication of the high quality of the material on each side of the boundary.

Finally, the angular dependence of the resistances within a grain and across the GB when a field of $\mu_0 H = 3$ T is rotated relative to the alignment of the *ab* planes of the sample at 89.3 K is shown in Fig. 3. Both sets of data show a pronounced decrease in resistance as the field is rotated towards the *ab* planes, consistent with the anisotropy of the material. The small distortion in the angular dependence of the boundary resistance when the field is close to the *ab* planes is likely to be due to a small *c*-axis misalignment between the grains which we have estimated independently, using optical microscopy, to be about 2°.

In conclusion, we have joined high quality quasisingle crystalline YBCO monoliths by a practical technique and characterized the electrical properties of the artificial GB thus created. The properties of the grains on either side of the boundary are comparable with the best values reported elsewhere. The IL and the angular dependence of the resistance of the boundary follows that of the grains very closely which suggests that the boundary critical current is determined by the same mechanism as in the grains and provides evidence of strong coupling across the boundary, even at fields of 5 T at 77 K. Although further work is required to elucidate the detailed form of the $E-J$ characteristics, and to obtain the critical current of the joined sample directly, the present results suggest strongly that these junctions are of high quality.

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