Direct Measurement of the Superconducting Properties of Single Grain Boundaries in $Y_1Ba_2Cu_3O_{7-\delta}$

P. Chaudhari, J. Mannhart, D. Dimos, C. C. Tsuei, J. Chi, M. M. Oprysko, and M. Scheuermann
IBM Research Division, Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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We have made the first direct measurements of the critical current density of individual grain boundaries and their adjoining grains in $Y_1Ba_2Cu_3O_{7-\delta}$ superconducting films. The critical current of the boundary was always found to be less than that of either of the two grains. The magnetic field dependence of the critical current of the grain boundaries indicates that they are comprised of regions of weak and strong Josephson coupling. A single grain boundary was also used to build and operate a simple dc SQUID. Current-voltage curves of a grain-boundary junction in the finite voltage range show subgap structure.

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There are a number of measurements on epitaxial thin films that show critical currents in the $Y_1Ba_2Cu_3O_{7-\delta}$ superconductors to be in excess of 10^5 A/cm^2 at 77 K.[1-4] In sharp contrast, polycrystalline films and bulk ceramic samples show substantially lower values.[5,6] This result has invariably been attributed to poor superconducting coupling at grain boundaries.[7-11] There has, however, been no direct demonstration that grain boundaries are indeed the cause of low critical currents. In this Letter, we report the results of our measurements of the electrical and magnetic properties of grain boundaries and of their two adjoining grains.

Thin polycrystalline films, approximately 0.5 μm in thickness, were deposited[10] onto polycrystalline $SrTiO_3$ substrates with large grains (approximately 100 μm or larger). After annealing, the films were epitaxially aligned with the grain orientation of the substrate resulting in a large-grained film. The grains were patterned with an excimer laser microfabrication technique[12] to make three lines. Two of these lines were within a grain and the third straddled the grain boundary. Samples were prepared with linewidths as small as 1 to 20 μm. The length of the line within a grain was approximately 40 μm whereas that at the grain boundary was between 2 and 40 μm. Electrical contacts were made with Al

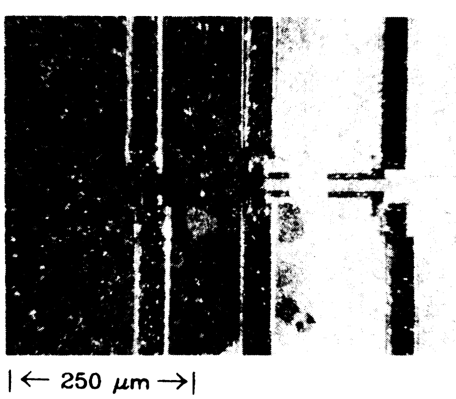

FIG. 1. Photograph of excimer-laser–patterned high-T_c film showing three isolated areas: two lines within adjacent grains and one line straddling the grains. With use of an oblique-incidence illumination technique, the two adjacent grains are clearly visible as dark and light area.

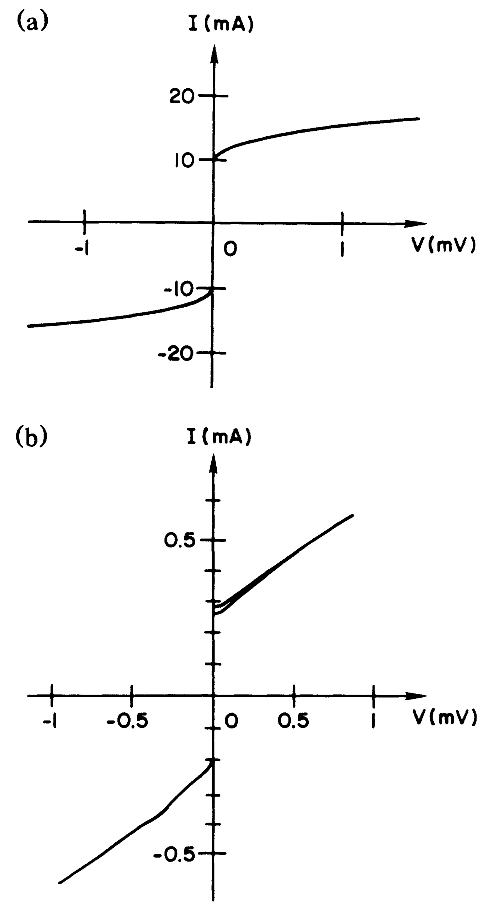

FIG. 2. Current-voltage characteristics of (a) an 8.5-μm-wide line inside a grain, and (b) a 4.5-μm-wide line straddling the boundary of the grain above and an adjacent grain.

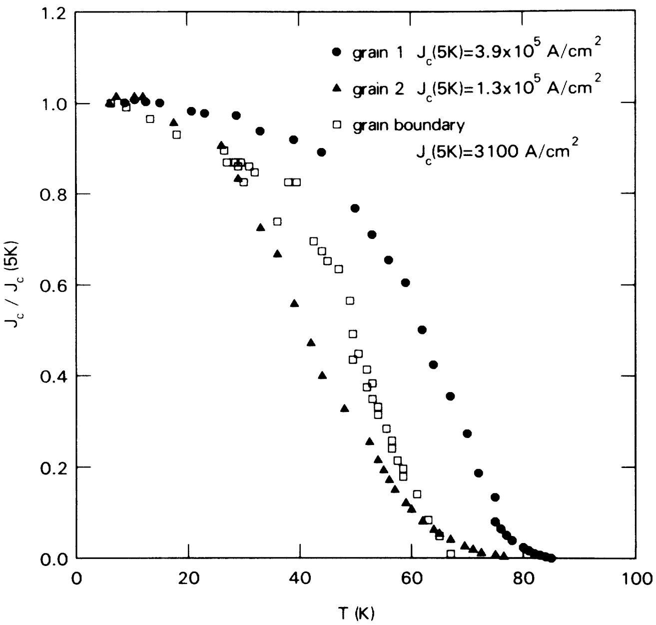

FIG. 3. Critical current density (normalized) vs temperature for the three lines in one sample.

wire bonds. An optical micrograph of a laser-patterned film is shown in Fig. 1.

Typical current-voltage characteristics measured with a four-point technique for a line within a grain and for one which crosses a boundary are shown in Fig. 2. The primary difference between their *I-V* characteristics close to zero voltage is that for the line crossing the grain boundary the onset of finite resistance showed steplike and frequently hysteretic behavior. The *I-V* curves are reminiscent of tunneling[13] between two superconductors with a small value of the Stewart-McCumber parameter.[14] The zero-voltage current, the critical current, for all three lines could be readily measured from the *I-V* curves and a typical plot of critical current versus temperature is shown in Fig. 3. The critical currents of the boundaries at 4.2 K ranged from 750 A/cm^2 in the worst case to values approaching 5×10^4 A/cm^2 in the best case. These values generally bracket the range of numbers reported widely in the literature on polycrystalline films and wires. Table I lists the critical current densities of the grains and their common grain boundaries. For the samples investigated to date the values of the critical current densities of the grains were always significantly larger than the line with the grain boundary. Hence our measurements would confirm the conjecture that grain boundaries limit the macroscopic critical currents of the bulk samples.

TABLE I. Critical current density (10^3 A/cm^2) at 4.2-5 K.

Grain 1	Grain 2	Grain boundary
> 500	> 500	50
> 100	70	18
250	220	12
390	130	3.1
260	14	0.7
8.5	5.8	0.7

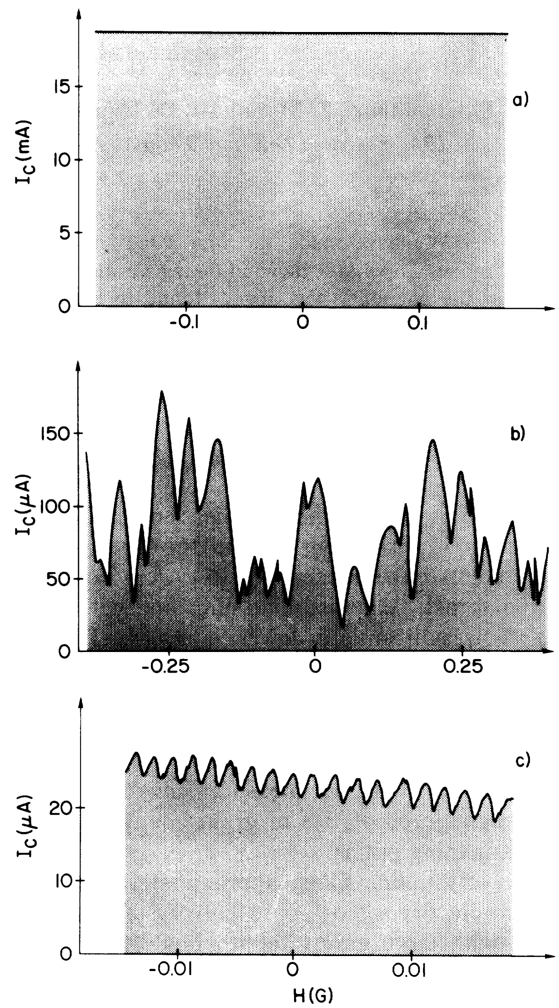

FIG. 4. Critical currents as functions of magnetic fields. (a) A 22-μm-wide line within a grain; (b) a 15-μm-wide line straddling a grain boundary; (c) a dc SQUID that was formed by two 8-μm-wide lines across another grain boundary. The effect of demagnetization has not been included in the computation of the actual field seen by the samples. The values quoted are the nominal fields generated by the magnet in the absence of a sample.

In order to understand further the behavior of the boundary, we have also investigated the *I-V* curves as functions of magnetic fields. Once again the response of the lines within a grain was quite different from that across the boundary. A typical plot of the critical

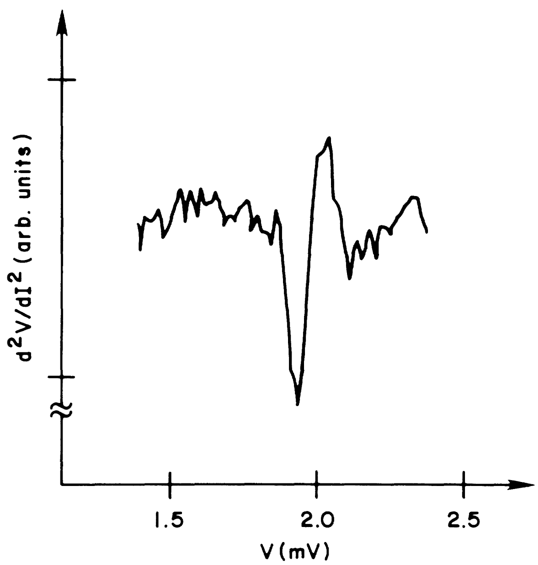

FIG. 5. Typical subgap feature in the d^2V/dI^2 as a function of voltage that was observed in lines crossing grain boundaries.

current as a function of the magnetic field is shown in Fig. 4. The field is applied parallel to the plane of the boundary and perpendicular to the plane of the film. The magnetic field dependence of the lines within a grain showed no features, which is the expected behavior. The behavior of lines containing a boundary show rich structure. This type of data suggests that a grain boundary is made up of regions of good and poor Josephson coupling. The interference pattern is a result of sampling of various wavelengths.

Recognizing that the superconducting coupling is relatively weak at the grain boundaries, we have built a dc SQUID structure in which the two junctions correspond to crossing of the same boundary twice, by drawing two laser-patterned lines across the boundary. As shown in Fig. 4(c) the expected periodic behavior with variation of applied field was observed. For purposes of comparison with Fig. 4(a) and 4(b), the full critical current dependence on magnetic field rather than the biased I-Φ characteristic usually presented in the literature is shown. The behavior of this SQUID is similar to the one reported on polycrystalline films in an earlier publication.[5]

The I-V curves of both the lines within a grain and those across a boundary were examined for structure in the finite voltage range. A variety of features in the first and second derivatives (dV/dI and d^2V/dI^2) versus voltage have been observed for the lines crossing grain boundaries. The principal difficulty with most of our samples is with our concern that flux flow in the lines might obscure intrinsic structure and energy scale. This requires preparation of samples in which the grains have very large critical currents. Below 2 mV we are confident that this is not a problem and hence we report that part of our data here. Typically we see anywhere from one to four subgap structures. They range in value from 40 μV to 2 mV. In two of the samples we observed multiple structure. One set had values at approximately 40, 80, 175, and 350 μV and the other at approximately 250, 500, and 1000 μV. We note that in these two sets the energy scale is approximately a multiple of the lowest value. An example of subgap structure is shown in Fig. 5.

In summary, we have shown by making direct measurements of laser-patterned regions that the superconducting coupling at grain boundaries is weak and varies spatially. The magnitudes of the critical currents we measure across the boundary are within the range of what is observed in polycrystalline films and bulk samples. The grain boundaries can be used to form SQUID structures and might provide a natural way to develop high-temperature Josephson devices which can be used for scientific or practical applications.

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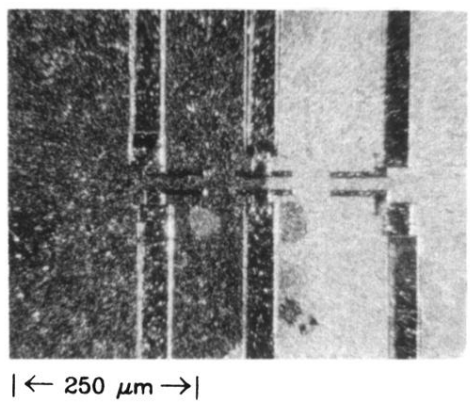

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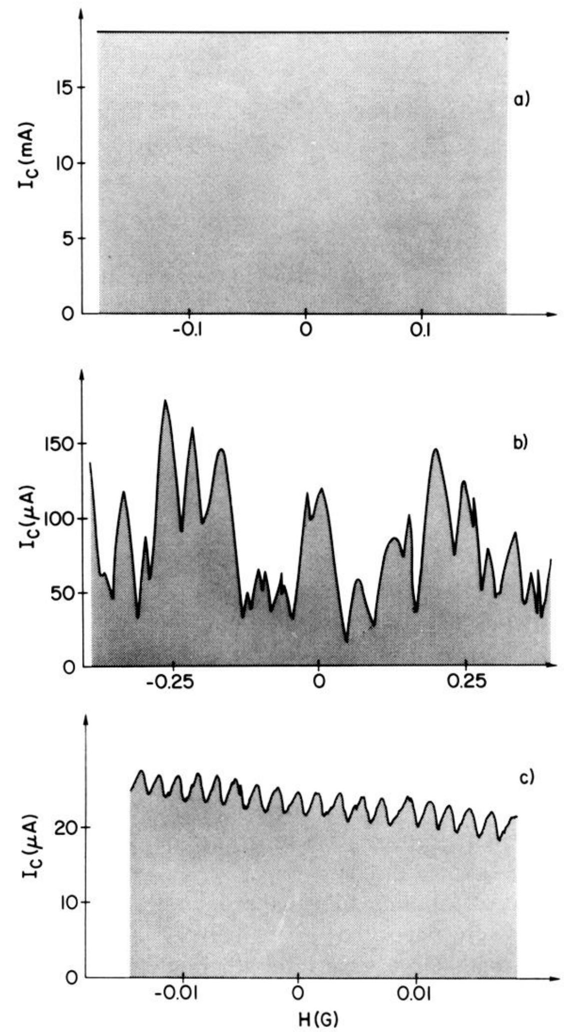

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