

# Evidence for weak link and anisotropy limitations on the transport critical current in bulk polycrystalline $Y_1Ba_2Cu_3O_x$

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Measurements of the transport critical-current density ( $J_c$ ), magnetization  $J_c$ , and magnetoresistance in a number of bulk sintered samples of  $Y_1Ba_2Cu_3O_x$  from several different laboratories indicate that the transport  $J_c$  is limited by weak-link regions between high  $J_c$  regions. The weak-link  $J_c$  has a Josephson character, decreasing by two orders of magnitude as the magnetic field is increased from 0.1 to 10 mT at 77 K. An examination of the grain-boundary region in  $Y_1Ba_2Cu_3O_x$  shows no observable impurities or second phases to the scale of the [001] lattice planes ( $\sim 12 \text{ \AA}$ ). The effect of intrinsic conduction anisotropy is discussed. A current-transfer model is proposed in which weak conduction along the  $c$  axis plays a role in limiting  $J_c$  at grain boundaries. Orienting the grains in the powder state during processing may result in enhanced transport  $J_c$  in bulk conductors.

## I. INTRODUCTION

Following the discovery of high-temperature superconductivity by Bednorz and Müller in metallic oxides,<sup>1</sup> a number of similar materials have been observed to have high transition temperatures, most notably the La-Sr-Cu-O system<sup>2-5</sup> with transition temperatures in the range 30–40 K, and the Y-Ba-Cu-O system<sup>6</sup> with transition temperatures above 90 K. Although the critical temperature  $T_c$  of these materials is high, their range of application will be determined by their ability to carry current. Most applications require high-current densities, usually more than  $10^4$ – $10^5$  A/cm<sup>2</sup>. Bulk applications usually further require high-current capacity and high  $J_c$  in the presence of a significant magnetic field (1–10 T). In *zero applied magnetic field*, the highest  $J_c$  reported<sup>7</sup> at 77 K for bulk polycrystalline  $Y_1Ba_2Cu_3O_x$  material is about  $10^3$  A/cm<sup>2</sup>, with  $J_c$  values in the range of 200 A/cm<sup>2</sup> being more usual.<sup>8</sup> This value of  $J_c$  is significantly lower than that needed for most bulk applications. In a magnetic field,  $J_c$  is much lower.

Although values of  $J_c$  are commonly calculated from magnetization data, very little data are available on directly measured values of the transport  $J_c$  for  $Y_1Ba_2Cu_3O_x$  in magnetic-fields. The first measurements of the magnetic-field dependence of the transport  $J_c$  in  $Y_1Ba_2Cu_3O_x$  were recently reported.<sup>8,9</sup>

This paper is based on those data along with additional data on transport  $J_c$ , magnetization  $J_c$ , strain effect on  $J_c$ , magnetoresistance, and transmission electron microscopy of the grain-boundary region in bulk polycrystalline  $Y_1Ba_2Cu_3O_x$ .

A number of bulk sintered polycrystalline samples from several different laboratories were tested. The data show

three characteristics which were common to all samples: (1) For  $T < T_c$  the transition of the resistance of the material from the normal-state value to zero occurs gradually over a very wide magnetic-field range. (2) The transport  $J_c$  drops sharply by two orders of magnitude between 0.1 and 10 mT. (3) The transport  $J_c$  is about two orders of magnitude lower than the  $J_c$  calculated from magnetization data under the assumption of a homogeneous current distribution.

These general characteristics are interpreted as evidence that the transport  $J_c$  in bulk polycrystalline  $Y_1Ba_2Cu_3O_x$  is dominated by weak-link regions separating high  $J_c$  regions in the material.<sup>8,10,11</sup> Possible causes of the weak-link phenomenon are considered, including impurities and compositional variations at the grain boundaries, and intrinsic anisotropy of the superconducting properties. Microscopic examination of the grain-boundary region of  $Y_1Ba_2Cu_3O_x$  by transmission electron microscopy (TEM) shows no observable impurities or second phase at the grain boundaries to the scale of the [001] lattice planes ( $\sim 12 \text{ \AA}$ ). X-ray energy-dispersive spectroscopy (XEDS), and electron-energy-loss spectroscopy (EELS) also show no changes in composition at the grain boundary, but to a coarser scale of about 200 Å. The microscopy data showing the mismatch of the Cu-O planes at the grain boundaries, plus the intrinsic anisotropy of  $J_c$  observed in single crystals,<sup>12</sup> indicates that one of the factors limiting  $J_c$  in polycrystalline material may be intrinsic conduction anisotropy. The process of current redistribution among the highly conducting Cu-O planes at grain boundaries will be limited by the intrinsic weak conduction perpendicular to the Cu-O planes. This suggests that orienting the grains in bulk materials would have the potential to realize an enhanced  $J_c$ , and may account for some of the difference in  $J_c$  between polycrystalline samples and epitaxial  $Y_1Ba_2Cu_3O_x$  films.<sup>13</sup> Some general guidelines for tech-

niques to accomplish this in bulk samples are outlined in Ref. 14.

## II. EXPERIMENT

Bulk sintered samples of  $Y_1Ba_2Cu_3O_x$  (where  $x \leq 7$ ) from three different laboratories were tested.<sup>9,15,16</sup> Preparation details on specific samples are given in Table I. Typically the samples were made from powders of  $CuO$ ,  $Y_2O_3$ , and  $BaCO_3$  which were weighed after drying and then reacted at high temperature, usually in an oxygen atmosphere. The materials were homogenized by repeated grinding, mixing, and heating in flowing oxygen. This was followed by cold pressing the powder into final form and sintering the sample at high temperature in flowing oxygen. The samples were then cooled and soaked in flowing oxygen at temperatures between about 400 and 500 °C.

As noted in Table I, grain sizes ranged from 10 to 50  $\mu m$ , and the distribution of grain sizes was quite wide. Samples were about 75%–95% dense, as listed in Table I. A scanning electron micrograph of the surface of sample 4 is shown in Fig. 1.

All samples were in the form of bars, 1–3 cm long, with a cross-sectional area of about 2 mm<sup>2</sup>. The samples were tested using a four-terminal technique in magnetic fields ranging from 0.1 mT to 19 T.

Considerable care was taken to ensure that the results were not affected by heating at the current contacts. A lower limit on the contact heating level needed to drive the sample normal in liquid nitrogen was determined from a sample with relatively resistive current contacts. All data obtained at 77 K which are reported here were obtained on samples where the contact heating was at least 100 times less than this lower limit.

$J_c$  measurements were also made at temperatures below 77 K. These data were obtained using a 13.5-T superconducting magnet and a variable-temperature gas-flow cryostat. Temperature control was maintained using a capacitance thermometer with a precision better than 0.1 K. Data are reported at these lower temperatures only for currents low enough that the heating levels were ten times less

than the power level where the  $V$ - $I$  curves showed heating effects as evidenced by irreversibility.

## III. RESULTS

### A. $V$ - $I$ characteristics and magnetoresistance $R(H)$

Figures 2 and 3 show the voltage versus current ( $V$ - $I$ ) characteristics for two of the samples measured at fields from 1 mT to 19 T at 77 K in liquid nitrogen. The striking feature about these characteristics is that the voltage did not rise abruptly to the normal resistance value as current was raised above the critical current  $I_c$ . At current levels well above the critical current (indicated by arrows along the abscissa) the  $V$ - $I$  characteristics were nearly linear and had a slope that was much less than the normal resistance of the sample at  $T_c$ . As magnetic field was increased, the slope increased toward the normal-resistance value. We estimate on the basis of the high-field data that fields over 30 T are required to completely reach the normal-resistance value, i.e., suppress all superconductivity. In contrast, very low fields, only a few tesla, were required to suppress the transport  $I_c$  as noted by the arrows along the abscissa in Figs. 2 and 3.

Thus, there is a very wide range of magnetic field over which the material transforms from the superconducting to the normal state. Figure 4 shows the magnetic-field dependence of the resistance at 77 K of two samples measured at current levels much greater than  $I_c$  (that is, where  $I_c$  is exceeded and the  $V$ - $I$  characteristic is linear as shown in Figs. 2 and 3). At a magnetic field of 20 T, the resistance is only about 40% of the normal resistance,  $R_n$ , measured in low magnetic field just above  $T_c$ . Figure 4 also shows that the decrease in resistance levels off at low field to a value of about 10% to 13% of  $R_n$ .

### B. Transport critical-current density versus magnet field, $J_c(H)$

Figure 5 shows a typical trace of the low voltage region of the  $V$ - $I$  characteristics below  $I_c$ . This curve was taken in zero applied magnetic field, but in the presence of the earth's field of about  $0.5 \times 10^{-4}$  T. The  $V$ - $I$  curve was reversible.

TABLE I. Sample characteristics.

	Samples 2,4	Sample 5	Samples 1,3
Heat treatment	Fire and premix twice Final treatment: 940 °C for 16 h in O <sub>2</sub> 700 °C for 16 h in O <sub>2</sub> slow cool in O <sub>2</sub>	Calcine at 950 °C 3 h to 400 °C 5 h to 975 °C hold for 5 h slow cool in O <sub>2</sub>	Grind and sinter three times 950 °C for 10 h in O <sub>2</sub> slow cool in O <sub>2</sub>
Average grain size	10 $\mu m$	20–50 $\mu m$	10 $\mu m$
Grain size distribution		wide	3–20 $\mu m$
Density	75%	90%–95%	75%
Superconductor cross-sectional area	Sample 2: 1.98 mm <sup>2</sup> Sample 4: 1.42 mm <sup>2</sup>	1.77 mm <sup>2</sup>	Sample 1: 0.74 mm <sup>2</sup> Sample 3: 2.11 mm <sup>2</sup>
$\rho(T_c)$	Sample 2: 595 $\mu\Omega$ cm Sample 4: 344 $\mu\Omega$ cm	1088 $\mu\Omega$ cm	Sample 1: 254 $\mu\Omega$ cm Sample 3: 317 $\mu\Omega$ cm

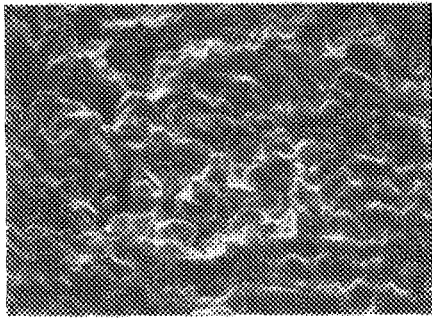


FIG. 1. Scanning electron micrograph of surface of sample 4 showing granular structure.

The data show that the voltage below the critical current is zero within experimental accuracy ( $\pm 50$  nV).

As magnetic field is increased, the onset of the upward curvature in the  $V$ - $I$  curve is shifted toward zero, as shown in Fig. 6. The onset reached zero current at a magnetic field of about 2–5 T. At higher fields the sample appears to have finite resistance at zero current. The measurement accuracy is limited at low-resistance levels because of the very low current, thus it is difficult to examine this region of the  $V$ - $I$  curve. However, there is a clear nonlinearity or offset in the  $V$ - $I$  characteristic even in fields as high as 10 T. Thus, there is some remnant of superconductivity evidenced by the transport current in fields of at least 10 T, although the current and resistance levels are far from any practical value.

The critical-current density measured in seven samples in the earth's magnetic field ranged from a low of about 10 A/cm<sup>2</sup> to a high of about 200 A/cm<sup>2</sup>. Thus, the transport critical-current densities were generally low and variable. No correlation between the critical current and the external-lead contact resistance was observed, confirming that contact heating at the current connections was not the cause of

the low  $J_c$  values. Also, as noted above, data are presented only on samples for which the contact heating was at least two orders of magnitude below the minimum contact heating level needed to drive the superconductor normal.

When magnetic field is applied perpendicular to the direction of transport current,  $J_c$  is suppressed significantly below the zero-field value. Figure 7 shows the magnetic-field dependence of  $J_c$  in two samples. The critical-current density for both samples decreased sharply by more than two orders of magnitude as magnetic field increased from 0.1 to 10 mT. The decrease in  $J_c$  with magnetic field was less rapid above 1 T, but in this region the  $J_c$ -vs-field plot is strongly dependent on the criterion used to determine  $J_c$ . (The higher  $J_c$  region below 1 T is not greatly affected by this criterion, however.) Two criteria are shown in Fig. 7. The solid curves correspond to an electric-field criterion of  $1 \mu\text{V}/\text{mm}$ , along with a correction for current carried by the sample when it is in the normal state. The dashed curves are for a resistivity criterion of  $1 \mu\Omega \text{ cm}$ . Only very coarse resistivity criteria could be applied to the data in the high-field region where the critical current is extremely low because of the limits on electric-field detection in such short samples. (At very low fields where  $J_c$  is much higher, the resistivity criteria can be correspondingly more sensitive.) Regardless of which criterion is used, the data show that the effective upper critical field for transport  $J_c$  is significantly less than 10 T. This is much less than the estimated 30-T field needed to completely suppress all superconductivity (i.e., where the resistance approaches the normal-state value).

The effect of stress on the critical current of samples 3, 4, and 5 was also measured at very low tensile strains up to about 0.05%. Higher strains caused the samples to fracture. No significant effect was observed, but, on the other hand, no measurable effect was expected at this low strain level. In Nb<sub>3</sub>Sn, for example, where the critical current is relatively sensitive to strain, the effect of 0.05% strain at low fields is to degrade the critical current by only 0.3%.<sup>17</sup>

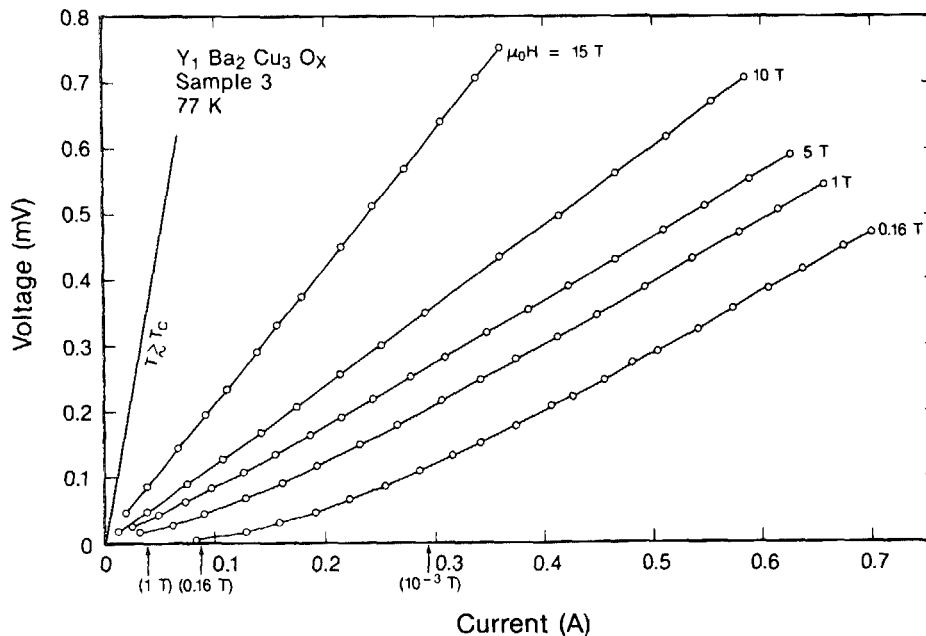


FIG. 2. Voltage vs current characteristics for sample 3 in transverse magnetic fields in liquid nitrogen at 77 K. At currents well above the critical current (indicated by arrows along the abscissa) the  $V$ - $I$  curves were nearly linear. High magnetic fields are required to increase the slope to the normal-resistance value at  $T_c$ , but the transport  $J_c$  is suppressed by very low fields.

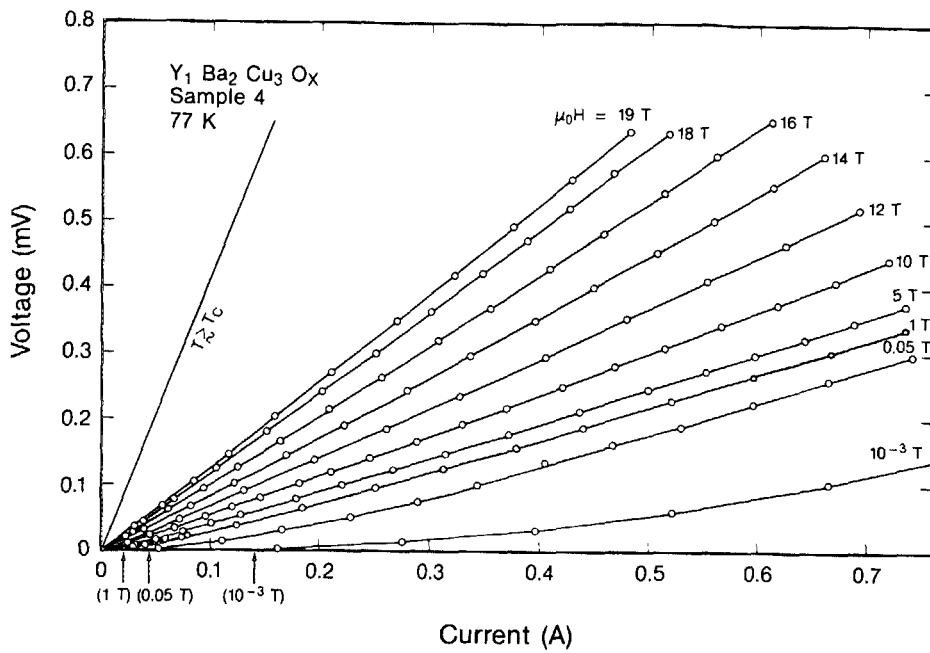


FIG. 3. Voltage vs current dependence for sample 4 in transverse magnetic fields showing results similar to Fig. 2 and the generality of the shape of the transport characteristics for different samples (after Ref. 8).

The characteristics reported here appear to be general among different samples made by different laboratories. All samples showed the same features of a linear  $V$ - $I$  curve above  $J_c$ , a high field required to totally suppress all superconductivity, and a low field required to suppress the critical current.

Data on  $J_c$  versus magnetic field at temperatures below 77 K are presented in Fig. 8.  $J_c$  was determined using an electric-field criterion of  $0.1 \mu\text{V}/\text{mm}$ . The data labeled " $H = 0$ " were taken in the residual field of the superconducting magnet ( $\sim 1 \text{ mT}$ ). This sample has a  $J_c$  of  $233 \text{ A}/\text{cm}^2$  in the earth's field and 77 K. The temperature dependence of  $J_c$  near  $T_c$  is shown in Fig. 9.

### C. Magnetization $J_c$ versus transport $J_c$

These values for the transport  $J_c$  appear significantly lower than values of  $J_c$  calculated from magnetization mea-

surements assuming that the induced current distribution is homogeneous.  $J_c$  has been calculated at fields above 0.5 T from magnetization data of Panson *et al.*<sup>15</sup> at 77 K on a material fabricated in the same way as sample 3 in Fig. 7. Values of magnetization  $J_c$  are shown as a dotted line in Fig. 7 at magnetic field between 0.5 and 6 T. A comparison with the transport  $J_c$  shown in Fig. 7 for sample 3 indicates that the magnetization  $J_c$  is about two orders of magnitude higher than the transport  $J_c$  over the same magnetic-field range.

### IV. EVIDENCE FOR WEAK-LINK COUPLING

These results indicate that the ranges of values of both the upper critical field and  $J_c$  in polycrystalline  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_x$  are very broad. More specifically, the data are consistent with a model in which the transport  $J_c$  is dominated by weak-link regions having very low  $J_c$ , separating high  $J_c$  regions in the material. The evidence offered by these data for such a hypothesis is described in this section.

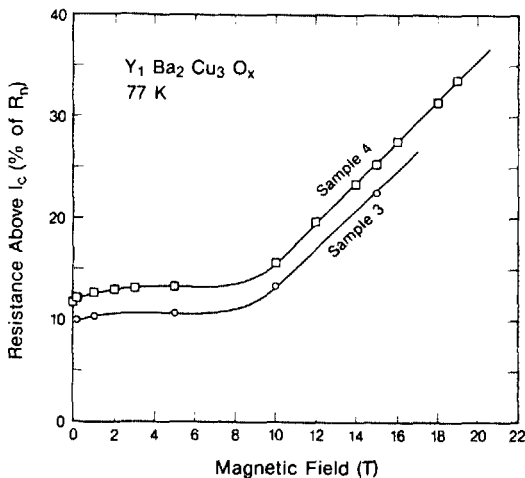


FIG. 4. Magnetic-field dependence of the resistance of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_x$  above  $J_c$ . The resistance is shown as a percentage of the normal-state resistance,  $R_n$ , measured at  $T_c$  in low magnetic field.

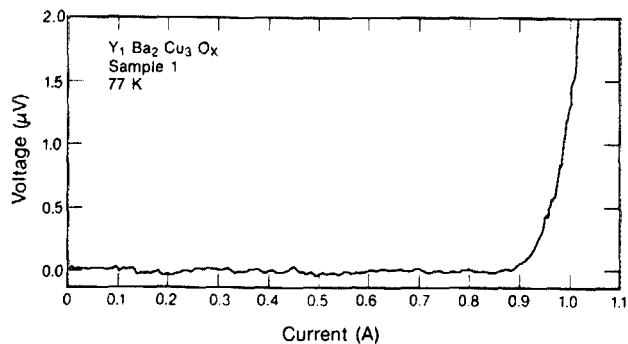


FIG. 5. Voltage vs current characteristic in earth's magnetic field ( $\sim 0.05 \text{ mT}$ ) showing the absence of detectable voltage below the critical current (after Ref. 8).

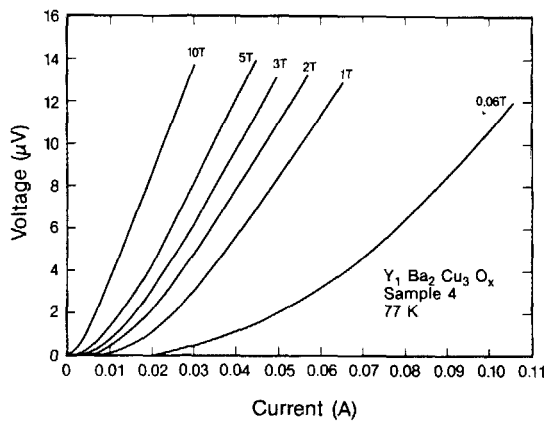


FIG. 6. Voltage vs current characteristics in applied transverse magnetic fields up to 10 T.

### A. $R(H)$

The very wide magnetic-field range over which the resistivity of the material increases from zero to the normal-state value is indicative of an inhomogeneous material with a wide range of  $H_{c2}$ . Figure 4 shows that at very low magnetic fields the resistance is nearly constant. As magnetic field is increased, there is a break in the curve at about 3 T and the resistance starts to increase toward the normal-state value. We speculate that this nearly constant low-field resistance is a regime wherein the transport current has driven the weak-link region normal, but the applied field is much less than  $H_{c2}$  for the high  $J_c$  regions. Assuming this is true, we find for these two samples that the weak-link region contributes a

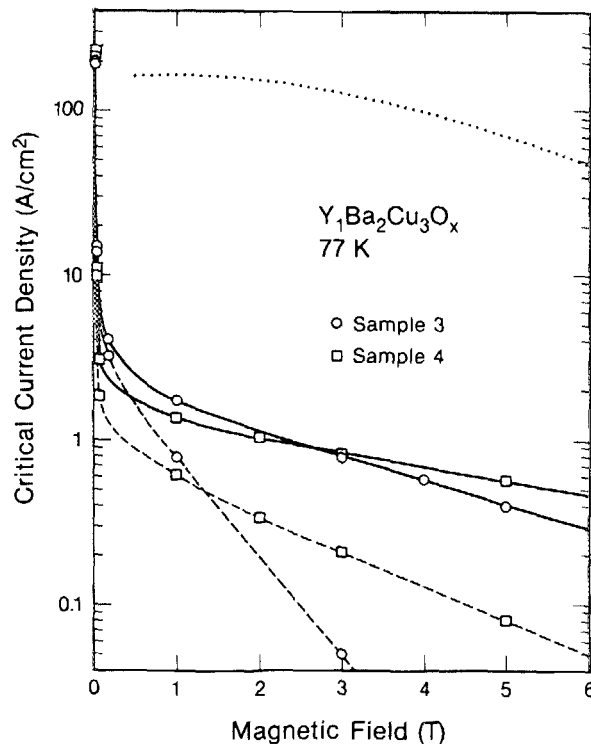


FIG. 7. Transport critical-current density  $J_c$  vs perpendicular magnetic field for polycrystalline  $Y_1Ba_2Cu_3O_x$  samples at 77 K. The solid line corresponds to an electric-field criterion of  $1 \mu V/mm$ ; the dashed line corresponds to a resistivity criterion of  $1 \mu \Omega cm$ ; the dotted line indicates the magnetization  $J_c$  measured in a sample similar to sample 3. The two lowest-field points were taken at 0.05 and 1 mT. There is a very sharp decrease in  $J_c$  in this low-field regime.

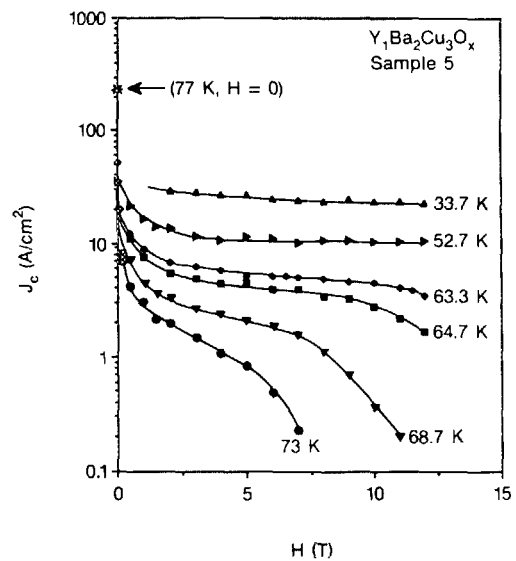


FIG. 8. Transport  $J_c$  vs magnetic field at temperatures below 77 K.

resistance equal to about 10%–13% of the normal-state resistance at  $T_c$ .

Under this interpretation, the constant resistance region of the  $R(H)$  curve is a result of high measuring current and should disappear at sufficiently low currents. Thus, care should be used in focusing on this part of the  $R$ -vs- $H$  curves to determine at what field the resistance reaches zero, since we suspect such results<sup>11,18</sup> are highly dependent on measuring current.

At higher fields around 8 T, where the  $R(H)$  curve starts to turn upward, the magnetic field (rather than measuring current) starts to suppress superconductivity in the high  $J_c$  regions of the material. Some idea of the minimum  $H_{c2}$  at 77 K can be obtained by extrapolating the high-field characteristic in Fig. 4 to zero resistance. A straight line extrapolation reaches zero at about 3 T. This would indicate that the minimum value of the range of  $H_{c2}$  at 77 K is quite low, on the order of 3–8 T. This is consistent with the field range where the onset of resistance at zero current is ob-

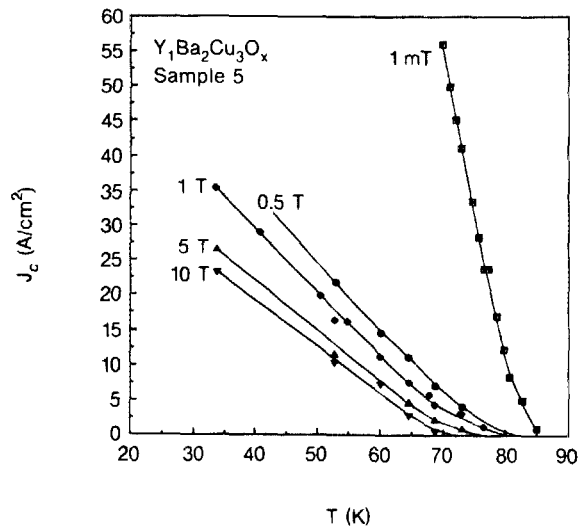


FIG. 9. Transport critical-current density  $J_c$  vs temperature for several values of applied magnetic field.

served in the critical-current curves of Fig. 6. The upper limit of the range of  $H_{c2}$  is much greater than 20 T, as indicated by the fact that the resistance reaches only 40% of the normal-state value at 20 T.

Thus, the data can be interpreted as evidence for a broad range of  $H_{c2}$  at 77 K extending from a minimum value of about 3 T to a maximum value much greater than 20 T. Such an interpretation is supported by recent data<sup>12</sup> showing anisotropic  $H_{c2}$  in single crystals of  $Y_1Ba_2Cu_3O_x$ . Recent calculations<sup>18,19</sup> show that the shape of the high-field portion of the  $R(H)$  curve can be explained at least in part by an anisotropic  $H_{c2}$  and percolation paths through randomly oriented crystallites. In addition to  $H_{c2}$  anisotropy, conduction anisotropy (discussed below) may also be a contributing factor in interpreting these  $R(H)$  data.

### B. $J_c(H)$

The magnetic-field dependence of  $J_c$  is consistent with a picture in which the transport  $J_c$  is dominated by weak-link regions having very low  $J_c$  that separate high  $J_c$  regions within the material. In such a situation, the low  $J_c$  regions would dominate the transport  $J_c$ . The  $J_c$  decreases by two orders of magnitude between 0.1 and 10 mT. This rapid decrease in  $J_c$  at very low fields is indicative of Josephson weak-link currents, which are suppressed by very low magnetic fields. (We are using "Josephson weak-link" as a generic term and do not mean to differentiate between SIS, SNS, or SS'S weak-link conduction.)

### C. Transport $J_c$ versus magnetization $J_c$

The concept of low  $J_c$  regions connecting high  $J_c$  regions is further supported by the large (two orders of magnitude) difference between  $J_c$  measured directly from transport currents and  $J_c$  calculated from magnetization data using the Bean model.<sup>20</sup> If weak-link regions were present, they would completely dominate the transport  $J_c$ , but only partially affect the magnetization  $J_c$ . This is easy to see in the extreme case where the high  $J_c$  regions are completely decoupled by the weak regions. In such case, the transport  $J_c$  would be zero, but circulating supercurrents within the high  $J_c$  regions would still give a finite magnetization and, hence, a finite magnetization  $J_c$ . Values of  $J_c$  calculated using the Bean model are applicable only to homogeneous superconductor materials, not highly heterogeneous materials such as these. Thus, the large difference observed between the magnetization  $J_c$  and the transport  $J_c$  is evidence for high and low  $J_c$  regions with the weak region dominating the transport  $J_c$ . This interpretation is also consistent with observations by magnetic susceptibility<sup>21</sup> of two superconducting components in Y-Ba-Cu-O.

The spatial extent of the weak-link region is uncertain. Since the material is granular, inhomogeneities in the structure might occur on a scale the size of an individual grain. An upper limit on the size can be obtained from observations such as those reported in Ref. 10 where hysteresis of the magnetization  $\Delta M$  is measured in a bulk sintered sample, and then the sample is powdered to near grain size and magnetization remeasured. There is not much change in the

measured magnetization. We see typically  $\Delta M_{\text{bulk}} / \Delta M_{\text{powder}} = 2-5$ . This suggests that the high  $J_c$  regions occupy a region no larger than a grain. On the other hand, scanning tunneling data<sup>22</sup> on single-crystal samples suggest that there may be "granularity" within each grain. That is, the tunneling  $I-V$  traces at low bias show a linear conductance indicative of Coulomb gaps brought about by domains on a scale less than  $\frac{1}{10}$  the grain size. Thus, the size scale of the heterogeneity is uncertain.

## V. ROLE OF CONDUCTION ANISOTROPY

In this section we consider some possible sources of the weak-link behavior in  $Y_1Ba_2Cu_3O_x$ . The evidence is based on microscopy data of the grain-boundary regions in  $Y_1Ba_2Cu_3O_x$  as well as recent data on epitaxial films and single-crystal  $Y_1Ba_2Cu_3O_x$  samples.

An obvious possibility is impurities or second-phase material at the grain boundaries. A transmission electron micrograph (TEM) lattice image of a grain-boundary region in bulk polycrystalline  $Y_1Ba_2Cu_3O_x$  is shown in Fig. 10. In this technique the diffraction conditions in the microscope are set up to image the [001] planes (parallel to the Cu-O planes) in adjacent  $Y_1Ba_2Cu_3O_x$  grains. Hence, the  $c$  axis of the orthorhombic structure is normal to the observed fringes. In all cases where grain boundaries are observed between 1:2:3 crystalline regions of the materials, clean grain boundaries such as shown in Fig. 10(b) are observed. The lattice image shows no observable impurities or second phases at the grain boundaries to the scale of the [001] lattice plane ( $\sim 12 \text{ \AA}$ ). X-ray energy-dispersive spectroscopy (XEDS) and electron-energy-loss spectroscopy (EELS) have confirmed that each grain is nominally 1:2:3 ( $Y_1Ba_2Cu_3O_x$ ) in metal composition to a resolution of about 200  $\text{\AA}$ . These results are also consistent with x-ray analysis on the La-Sr-Cu-O system where no change in composition near the grain boundary was observed.<sup>23</sup> This implies that if impurities, second phases, or compositional variations are a factor at the grain boundaries, they would have to be acting on a scale finer than that observed. A significant effect on such a fine scale cannot be ruled out, however,

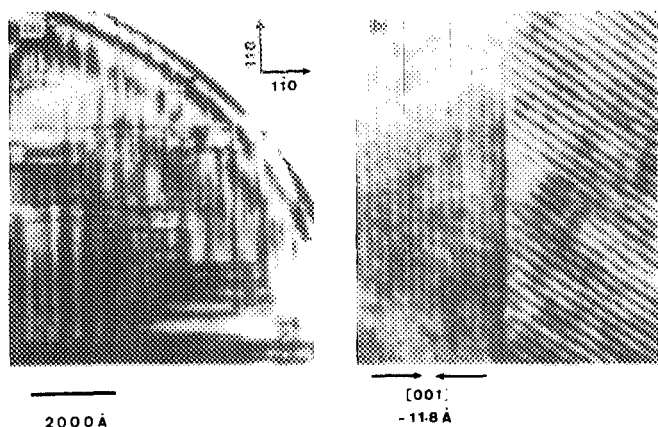


FIG. 10. (a) Low magnification TEM micrograph of twin boundaries within a grain of  $Y_1Ba_2Cu_3O_x$ . (b) High-resolution TEM of the boundary region between grains obtained using a lattice imaging technique. There is a misalignment of the Cu-O atomic planes at the grain boundary in polycrystalline  $Y_1Ba_2Cu_3O_x$ .

because of the very small coherence length in these materials. Higher-resolution analysis are needed, particularly of possible compositional variations near grain boundaries.

There are regions of all samples (<20% by volume) which consist of either amorphous or fine-grained ( $\sim 20 \text{ \AA}$ ) polycrystalline material possessing distinctly different compositions than 1:2:3. However, the amount of this type of material is insufficient to produce a lack of percolation between the 1:2:3 crystalline materials.

We now consider the inherent anisotropy of the superconducting properties of the  $Y_1Ba_2Cu_3O_x$  crystal structure as a possible mechanism contributing to the low  $J_c$  in polycrystalline  $Y_1Ba_2Cu_3O_x$ . Recent data<sup>12</sup> on single-crystal samples of  $Y_1Ba_2Cu_3O_x$  show that there is a strong anisotropy in the magnetization  $J_c$  within the  $Y_1Ba_2Cu_3O_x$  crystal structure, with conduction in the plane of the Cu-O chains being higher than in the direction perpendicular to these planes. For example, at 40 K in a field of 1 T, the  $J_c$  anisotropy between conduction parallel and perpendicular to the Cu-O planes in one sample was more than 500 and increased rapidly with field. Other samples were reported to show anisotropy several times larger.<sup>12</sup> (This is a lower bound on the  $J_c$  anisotropy, since the anisotropy was reported to decrease more than an order of magnitude at 2 T for a 5° misalignment of the crystal.) The upper critical field has been observed to have significant anisotropy as well.<sup>12,24</sup>

The magnetic-field dependence of  $J_c$  obtained from magnetization data<sup>12</sup> in single crystals also shows a much sharper fall off for  $J_c$  perpendicular to the Cu-O planes (the *c*-axis direction) than parallel, decreasing more than two orders of magnitude between 0 and 1 T at 40 K. At 77 K, no single-crystal data were available, but judging from the trend, we expect the decrease with magnetic field to be greater at higher temperatures. These single-crystal results indicate that at least part of the sharp decrease of  $J_c$  with magnetic field in bulk samples at higher temperatures may arise from the intrinsic properties of weak conduction in the *c*-axis direction.

The steep magnetic-field dependence indicates that conduction along the *c* axis may have a Josephson weak-link character<sup>25</sup> with a steep magnetic-field dependence at low field. Recent band-structure calculations<sup>26</sup> indicate a two-dimensional Fermi surface with a charge deficit in the vicinity of the rare-earth planes, so there is some physical basis for expecting such a weak-link conduction character in the *c*-axis direction. Alternatively, weak flux pinning for current conduction in this direction may also be a factor contributing to the steep magnetic-field dependence of  $J_c$ .

In either event these data are consistent with *intrinsic conduction anisotropy* within the crystallites being a significant factor limiting the transport  $J_c$  in polycrystalline samples. At grain boundaries between randomly oriented crystallites, there is a situation where the highly conducting Cu-O planes are not in a one-to-one registry. The percolation conductivity will be limited by currents redistributing between the Cu-O planes in the vicinity of each grain boundary. This current-transfer process is limited by the intrinsic weak conduction perpendicular to the Cu-O planes, which could account for at least part of the great difference in  $J_c$

between polycrystalline samples and epitaxial thin films<sup>13</sup>.

This current-transfer model would suggest that the anisotropy limitation on transport  $J_c$  would be significantly greater at polycrystalline grain boundaries than at twin boundaries, since in the case of twinning, there is symmetry across the twin boundary, with a one-to-one correspondence of Cu-O planes on either side. However, in polycrystalline materials, there is a situation such as shown in Fig. 10(b), where many Cu-O planes on the right-hand side of the grain boundary would conduct current into a single Cu-O plane on the left-hand side of the grain boundary. This single plane cannot carry all the current introduced into it and so current is forced to transfer in the weak direction to adjacent Cu-O planes to equalize the current distribution. The geometry is similar to the calculation of current transfer in conventional multifilamentary superconductors,<sup>27</sup> except that here we are considering current transfer between planes on an atomic level as shown in Fig. 10(b), rather than macroscopic filaments.

Different approaches to production of these materials may be necessary for practical bulk applications of  $Y_1Ba_2Cu_3O_x$  in light of the anisotropy of the superconducting properties of these materials. In particular, if anisotropy is playing a key role, methods may have to be developed for orienting the crystallites in polycrystalline materials to enhance  $J_c$ . This may be accomplished by using a substrate to define a preferred crystal direction in the grains, as was done for the epitaxially grown films of  $Y_1Ba_2Cu_3O_x$  described above.<sup>13</sup> More economical techniques suited to bulk high-current conductors might also be possible, such as orienting the grains in the powder state during conductor fabrication.<sup>14</sup>

From the practical standpoint of magnet design, the best orientation would be to have the weak conduction *c* axis of the  $Y_1Ba_2Cu_3O_x$  crystal structure oriented in a direction mutually perpendicular to both the current and field directions. This provides both high critical current and high upper critical field, since the critical field is greatest for magnetic field parallel to the copper-oxygen planes.<sup>12</sup>

## VI. CONCLUSIONS

Bulk polycrystalline  $Y_1Ba_2Cu_3O_x$  materials are characterized by extreme heterogeneity in their transport properties. This is evidenced by the broad magnetic-field range of the superconducting transition, the sharp decrease in  $J_c$  at very low magnetic field, and the significant difference between transport  $J_c$  and magnetization  $J_c$ . These data also indicate that the transport  $J_c$  is characterized by high  $J_c$  regions separated by weak-link regions that have a Josephson weak-link conduction behavior.

TEM lattice images showing the mismatch of the Cu-O planes at the grain boundaries, along with recent anisotropy data in epitaxial films and single crystals, indicate that at least part of the limitation on  $J_c$  in bulk polycrystalline samples may result from intrinsic conduction anisotropy. The percolation process between randomly oriented grains in bulk samples necessitates a redistribution of currents between the Cu-O planes at grain boundaries. This current-transfer process will be determined by intrinsic weak con-

duction between the Cu-O planes when they are not in a one-to-one registry at the grain boundaries, such as would occur in bulk polycrystalline samples.

Different approaches to production of bulk  $Y_1Ba_2Cu_3O_x$  materials may be necessary. If anisotropy is playing a significant role, orienting the grains in the powder state during processing so that the weak conduction  $c$  axis is mutually perpendicular to the current and field directions may result in enhanced  $J_c$  and  $H_{c2}$  in bulk  $Y_1Ba_2Cu_3O_x$ .

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