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Josephson-junction model of critical current in granular $Y_1Ba_2Cu_3O_{7-\delta}$ superconductors

R. L. Peterson and J. W. Ekin

Electromagnetic Technology Division, National Bureau of Standards, Boulder, Colorado 80303

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We calculate the transport critical-current density in a granular superconductor in magnetic fields below about 5×10^{-3} T. The field dependence in this region is assumed to be controlled by intragranular or intergranular Josephson junctions. Various model calculations are fitted to transport critical-current data on bulk $Y_1Ba_2Cu_3O_{7-\delta}$ ceramic superconductors, whose average grain size somewhat exceeds 10 μm. The results yield an average junction cross-sectional area (thickness×length) of 4–6 μm^2. If the junctions are at the grain boundaries, a London penetration depth of about 150–300 nm is inferred, consistent with other estimates. We conclude that Josephson junctions are limiting the transport critical current in these samples and that they lie at the grain boundaries. The parameters of the fit are not consistent with Josephson junctions at twinning boundaries.

I. INTRODUCTION

A large decrease in the transport critical-current density J_c at low magnetic field in bulk sintered samples of $Y_1Ba_2Cu_3O_{7-\delta}$ (YBCO) was first reported by Ekin *et al.*[1] and Capone and Flandermeyer.[2] Similar observations have been made at several laboratories on other bulk sintered samples of YBCO. All show a large decrease (two orders of magnitude) of J_c at very low fields (10^{-4} to 10^{-2} T).[3-5] Further observations by Dinger, Worthington, Gallagher, and Sandstrom[6] have shown that large conduction anisotropy (three orders of magnitude) exists in single-crystal samples at fields above 1 T and temperatures above abut 40 K. These results have led to the suggestion by Ekin and co-workers[1,3,4] that the very low J_c in bulk materials is a result of Josephson weak-link effects in the low-field regime as well as an additional anisotropy limitation at higher fields and temperatures. The weak-link model for the low-field limitation is also strongly suggested by magnetization data reported by Suenaga *et al.*[7] and Larbalestier *et al.*[8] showing that J_c at fields less than 1 T is very high ($>10^6$ A/cm^2) within regions considerably smaller than the grain size. This interpretation is also consistent with observations of magnetic ac susceptibility as a function of temperature.[9,10]

The location of the weak links has been the subject of considerable speculation. Most investigators have suggested that the weak-link behavior dominant at low fields could be the result of Josephson junctions at the grain boundaries. Deutscher and Müller,[11] on the basis of some theoretical work and magnetization measurements, have argued that Josephson junctions exist *within* the grains of bulk YBCO, probably at twinning boundaries. Many have suggested that because the coherence length along the c axis is greater than the spacing between the Cu-O planes, there should be significant Josephson coupling between those planes.

In this paper, we present results which suggest that a choice can be made among these alternatives for current transport in bulk ceramic samples of YBCO. Our quantitative results strongly suggest that Josephson junctions indeed limit the transport current at low values of magnetic field; the measured value of their areas allows us to conclude that the junctions likely reside at the grain boundaries rather than at the twinning boundaries.

II. SAMPLE PREPARATION AND MEASUREMENT

Bulk sintered samples of $Y_1Ba_2Cu_3O_{7-\delta}$ with $\delta>0$ were made in the usual way as described in Refs. 4 and 12. 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 at 940 °C for 16 h in flowing oxygen, followed by an oxygen soak at 700 °C for 16 h and slow cooling in oxygen. Samples were about 75% dense. A scanning electron micrograph of the surface of one sample is shown in Fig. 1. This picture and others show that the average grain size is somewhat larger than 10 μm and that the distribu-

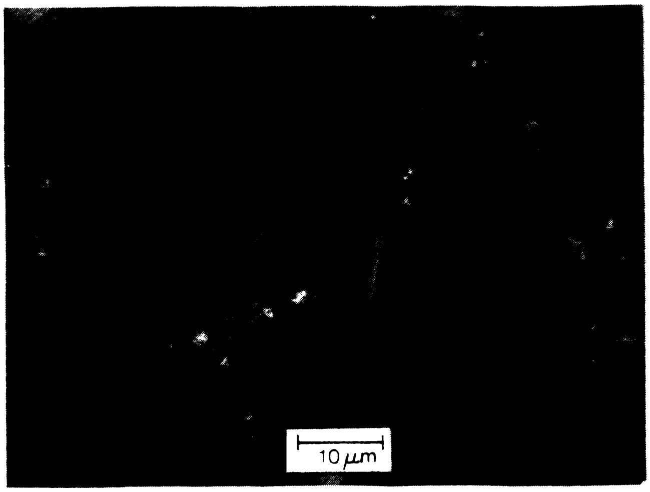

FIG. 1. A scanning electron micrograph of one of the samples studied. Individual grains and bands of twins can be seen.

tion of grain sizes is quite wide. The micrograph shows bands of twins; on the scale shown, individual twinning boundaries are not visible. A high-resolution micrograph on a similar sample[4] shows an average spacing of a few tens of nanometers between twinning boundaries, consistent with other reports.[13,14]

The critical-current measurements were carried out on samples cut into bars about 12 mm long, with a cross-sectional area of about 2 mm^2. The samples were tested using a standard four-terminal technique, with liquid nitrogen as a refrigerant. A cryostat was constructed with a mumetal shield to exclude the earth's magnetic field so that measurements could be made accurately down to magnetic fields of 10^{-5} T. The applied magnetic field was supplied by a copper solenoid and the system was calibrated with a Hall probe.

Considerable care was taken to ensure that the results were not affected by heating at the current contacts. Several samples were tested with contact resistivities differing by three orders of magnitude. The results were independent of the contact heating levels, indicating sufficiently low contact resistances even for the samples with higher contact resistance.

III. ANALYSIS

The starting point for our analysis is the assumption that the principal barrier to current flow in these granular superconductors, at fields below about 5×10^{-3} T, is a collection of Josephson junctions of random orientations and sizes. We assume that these junctions can be described by the usual Josephson equations. A magnetic field penetrating the barrier of a Josephson junction reduces the tunneling current.[15] If the junction length L is less than the Josephson penetration length λ_J, where

$$\lambda_J^2 = \frac{\Phi_0}{2\pi\mu d J_c(0)} ,$$

the current distribution within the junction is substantially uniform, and the critical current versus field relation is given by

$$J_c(H) = J_c(0)\left|\frac{\sin \pi H/H_0}{\pi H/H_0}\right| , \tag{1}$$

where the characteristic field H_0 is

$$H_0 = \frac{\Phi_0}{\mu d L} . \tag{2}$$

Here μ is the permeability and $d = 2\lambda + t$ is the effective junction thickness, where λ is the London penetration depth, and t is the barrier thickness. Figure 2 is a schematic representation of a Josephson junction with these parameters illustrated. The magnetic field extends into the superconductor a distance equal to about λ. If we take $d \approx 400$ nm and $J_c(0) \approx 200$ A/cm^2, as inferred from the present measurements, then $\lambda_J \approx 18$ μm. Since the average value of L would be about equal to or less than the average grain size, which is about 10–15 μm in the samples measured here, the condition $L < \lambda_J$ for the validity of Eq. (1) is satisfied.[16]

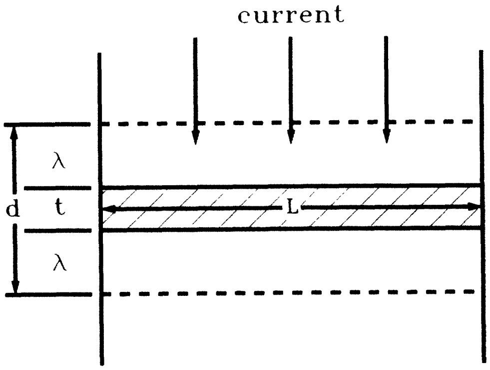

FIG. 2. A schematic representation of a Josephson junction between regions of the superconductor, showing the barrier thickness, the London penetration depths, junction length, and direction of net current. The effective component of magnetic field is normal to the plane of the paper.

Equation (1), which is a Fraunhofer diffraction pattern, has been used by Kwak, Venturini, Ginley, and Fu[5] in an estimated fit to their transport critical-current data on granular YBCO, without averaging over the junction lengths or orientations. Each junction will pass current according to its own Fraunhofer pattern, and interference among these patterns will smear out the structure. The dominant junction variable governing this smearing is the junction area, as our calculations show; the orientation of the junction relative to the magnetic field is less important.

We have performed averages over junction lengths using three different statistical distributions (rectangular, triangular, and skewed Gaussian), with various mean values and widths. The results [i.e., the estimates of H_0 defined in Eq. (2)] are found to be relatively insensitive to the type of distribution and the width of the distribution over a reasonable range of widths. For widths greater than about 50% of the mean of the distribution, the Fraunhofer structure becomes indistinct.

The effective magnetic field penetrating the junction is $H\sin\theta$, where θ is the angle between H and the normal to the plane of the junction, that is, the twinning plane or the surface of the grain at which the junction is located. In averaging over orientations, we assume that all θ are equally probable within a specified range (see the next paragraph). We find that a significant amount of Fraunhofer structure remains even when averaging over the full 90°, if an average over junction lengths is not made.

We have also considered the concept that in a precolative flow the least favorable orientations are the ones which control the current, and that the angular averaging is to be taken only over certain values of θ, presumably just near 90°. This introduces a new parameter, but we find that the results (the inferred values of H_0) are not very sensitive to the integration range.

The fact that the inferred values of H_0 are not very sensitive to the type and width of the distribution over lengths, and to the range of angles considered, gives confidence in the validity of our results.

The averaged value of $J_c(H)$ thus may be evaluated as

$$\frac{J_c(H)}{J_c(0)} = \frac{L_m}{\pi/2-\Theta}\int_0^\infty dx\,p(x)\int_\Theta^{\pi/2} d\theta \left|\frac{\sin(yx\sin\theta)}{(yx\sin\theta)}\right| , \tag{3}$$

where Θ defines the range of integration over θ, $x = L/L_m$, $y = \pi H/H_0$, and $p(x)$ is the probability distribution over lengths, normalized to unity. L_m is one of the parameters of the length distribution, and H_0 is now defined relative to L_m through Eq. (2), and is so used in Figs. 3 and 4.

In Fig. 3 we show the calculated critical current versus field together with data taken on two superconductor samples with similar grain size distributions. An electric field criterion of 1 μV/mm was used to determine J_c. For $J_c(H)/J_c(0) > 0.5$, the data were not very sensitive (less than 10%) to the particular criterion used. The sensitivity to the criterion used increases as J_c decreases. In Fig. 4 we show the same data and calculation on a log-log plot. The curves are the result of the double average: We integrate on θ from 80° to 90°, and we assume a skewed triangular distribution over lengths with $L(\text{smallest}) = 1$, $L(\text{peak}) = 7$, and $L(\text{largest}) = 22$ μm. The average length for this distribution is 10 μm. In the figures, we have chosen $\mu H_0 = \Phi_0/dL(\text{peak}) = 6\times10^{-4}$ T. The fit is good. For this case, we find $dL_{\text{av}} = 4.9$ μm^2. A calculation with the same length parameters but with $\Theta = 50°$ yields a curve shifted to the right by about a linewidth in Fig. 4. A symmetric length distribution with width equal to the mean gives a curve quite close to that of Fig. 4, with slightly smaller $J_c(H)/J_c(0)$ at the higher values of H. Broad distributions give the best fits. From fitting with different distributions, widths, and angle ranges, we estimate that the range for the junction cross-sectional areas dL_{av} is 4–6 μm^2.

If we take L_{av} to be 10 μm, we thus find the penetration depth to be 200–300 nm, assuming that the barrier thickness t is negligible compared to the penetration depth. This range of penetration depths is consistent with values obtained from other measurements.[12,17,18] Kwak *et al.*,[5] without averaging over lengths or orientations, estimated a London penetration depth of 280 nm from their transport critical current measurements, although it is not clear what value of junction length was used.

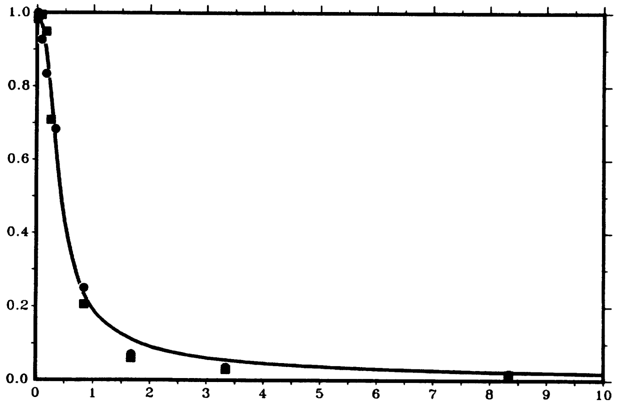

FIG. 3. Normalized transport critical current density vs applied field normalized to a characteristic field H_0. The curve is calculated for a skewed triangular distribution of junction lengths, and is averaged also over a range of angles (see text). The points are data taken on two samples of comparable grain size distributions, and are fit with the choice $\mu H_0 = 6.0\times10^{-4}$ T.

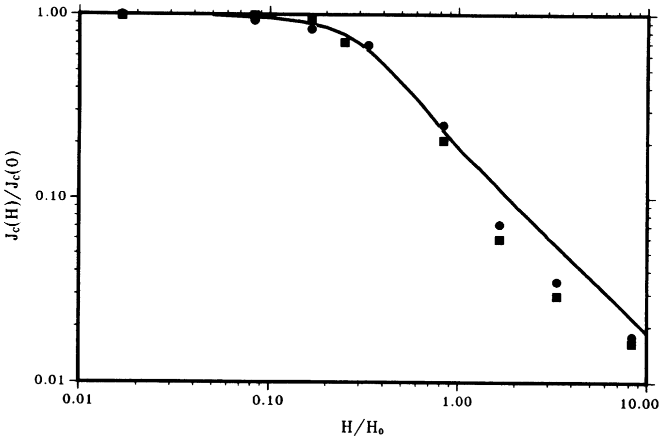

FIG. 4. The same information as in Fig. 3, plotted on a log-log scale.

Since by assumption the average length of the junctions is initially unknown, we should instead select a reasonable value for λ and deduce the size of L_{av}. For $\lambda = 200$ nm,[12] we find that L_{av} lies in the range 10–15 μm. This range is consistent with the average grain sizes in the samples studied.

IV. CONCLUSIONS

We thus are able to draw the following conclusions.

(1) Comparison of the theoretical with the experimental curves of transport critical current versus field gives convincing evidence that the impediments to transport current are indeed Josephson junctions.

(2) We have been able to deduce that the effective average junction area presented to a magnetic field is about 4–6 μm^2 in these samples.

(3) If the average junction length is about equal to the average grain size, the implied penetration depth is about 150–300 μm, consistent with other measurements. Conversely, if we accept 200 nm as a reasonable value for the London penetration depth, we infer an average junction length of 10–15 μm, comparable to the average grain size.

(4) The twinning boundaries in these samples are irregularly spaced with an average spacing of a few tens of nanometers. If these boundaries are Josephson junctions, their thicknesses (2λ) would be about 400 nm, overlapping many adjacent twinning planes. The entire interior of the grains would be penetrated rather uniformly by a magnetic field, and a very small dc magnetization would be observed. However, all the observations show a large response. To avoid a uniform internal magnetic field, one would have to postulate a λ of less than about 10 nm. But, when combined with our deduced junction area of 4–6

μm^2, this would demand a junction length more than ten times greater than the average grain size. The twinning boundary length is expected to be considerably less than the average grain boundary length. Our deduced junction area could then be recovered by postulating a much greater effective junction thickness than $2\lambda = 400$ nm, but such a procedure would be quite arbitrary, and again runs up against the uniform field problem mentioned above. Thus our results argue against Josephson junctions existing at the twinning boundaries.

(5) Josephson junctions at other defects within the grains are also not consistent with the parameters of the fit since their dimensions would be significantly less than the grain size.

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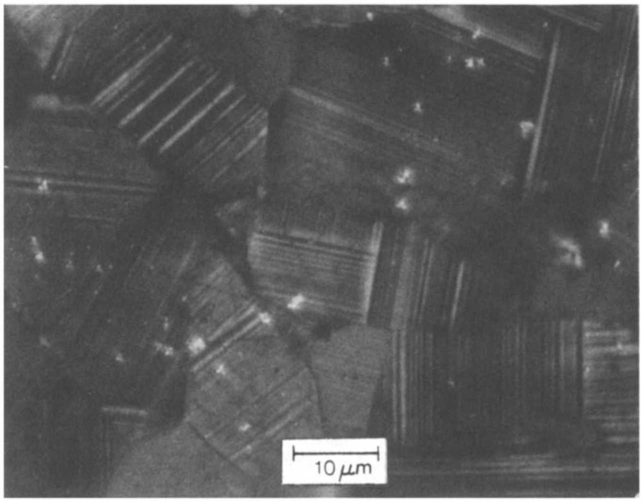

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