Temperature dependence of the critical current in high-T_c superconductors

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We present a study of the temperature dependence of the critical current in bulk samples of the high-T_c superconductors Y-Ba-Cu-O and Dy-Ba-Cu-O. We have found that the critical current varies linearly with $(T_c - T)$. This result suggests a percolating network of Josephson-coupled superconducting grains. However, the critical current versus temperature curves are found to have a nonlinear tail close to the critical temperature. We have determined that the temperature at which the I_c vs T curves depart from the linear behavior in our samples agrees with what is expected according to theoretical models of granular superconductors.

The recent discovery of high-T_c superconductors,[1,2] has generated a remarkable amount of activity in the study of the physical properties of these materials. The interest is twofold: From a practical point of view one would like to improve these materials and optimize those properties which are important in applications. To this end, a number of groups have measured the critical current densities that can be achieved in bulk,[3] thin-film,[4] and single-crystal samples[5] and how these critical current densities are modified by the presence of a field. From a purely scientific point of view, the challenge is to understand the mechanism which causes superconductivity in these materials, and to determine whether one can use those descriptions that have been so successful in describing low-T_c superconductors or if modifications to these approaches will be required in order to properly describe these new materials. Thus, it is of importance to study various properties of these materials and to attempt to understand them in the context of the usual phenomenological theories for superconductors including the necessary modifications to take into account the peculiarities of these materials such as their short coherence length, their anisotropy, as well as their percolative morphology. One of such properties which is well understood in conventional superconductors is the temperature dependence of the critical current density. While brief accounts of this variation are reported in the literature,[6] and specific predictions have been made of its temperature dependence,[7,8] a thorough comparison with experiment is still lacking.

In this paper, we present a detailed study of the temperature dependence of the critical current in Y-Ba-Cu-O and Dy-Ba-Cu-O compounds. We show that for homogeneous, single-phased samples the linear dependence of the critical current with temperature is consistent with a percolative network of grains coupled to each other through Josephson coupling. In one of our less homogeneous samples the critical current has two clearly different regimes which we interpret as the effect of the existence of two phases with different critical temperatures. In all cases, the Ginzburg-Landau description seems to remain valid in these materials.

Our samples were prepared under a variety of conditions. We consider a sample to be of good quality only if it exhibits a resistivity ratio bigger than one and low resistivity ($< 1000\ \mu\Omega\,\text{cm}$) at room temperature. As it is now known, we find the annealing sequence at the end of the sample preparation to be the most critical step during the fabrication, which apparently maximizes the occupancy of the 0, $\frac{1}{2}$, 0 oxygen sites in the structure. Our samples are made by the solid-state reaction of yttrium oxide, barium carbonate, and copper oxide in a furnace with continuous oxygen flow. The different compounds are mixed thoroughly in the desired proportions (in this paper we shall be concerned only with $R\text{Ba}_2\text{Cu}_3\text{O}_y$, R=Y and Dy). Then, the standard grinding-sintering-pressing procedure is followed. The crucial phase is the correct incorporation of oxygen into the structure. For our best samples this is done by reducing the temperature in steps of 200 °C for a total of two hours of annealing time in flowing oxygen.

The measurements were performed in a closed-cycle refrigerator with the sample bonded to a sapphire substrate by indium contacts. A four-terminal configuration was used in our measurements. Figure 1 shows a R vs T curve of a good sample. This sample has a resistivity ratio of 2.8 between room temperature and T_{onset}, the temperature at which the resistance starts to drop drastically.

Figure 2 shows the temperature dependence of the critical current for a sample without the careful cooling at the end of the fabrication process and with nominal composition $\text{Y}_{1.2}\text{Ba}_{0.8}\text{Cu}_2\text{O}_x$. This sample has a room-temperature resistivity ten times bigger than the rest of the samples reported in this paper and it has a low-resistivity ratio and, from the microscopic analysis, was found to have many phases as well as a coarse grain structure. There are two distinct regimes to the curve. Superconductivity is first detected at 82 K and the critical current grows

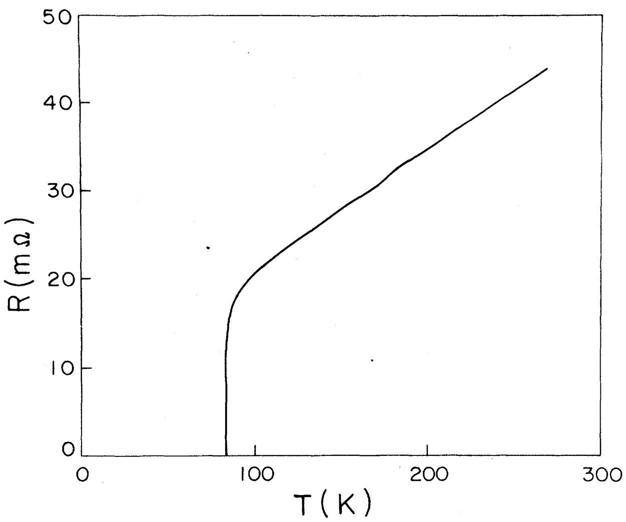

FIG. 1. Resistive transition of one of our best samples with nominal composition $YBa_2Cu_3O_y$. This sample has a high resistivity ratio, low room-temperature resistivity, and a transition width of 3 K.

linearly with (80 K $-T$) for 75 K $< T <$ 80 K. Below 75 K, the critical current grows linearly with (75 K $-T$) up to the lowest temperature measured ($\approx$50 K). This behavior can be seen more clearly in the logarithmic plots shown in the inset of Fig. 2.

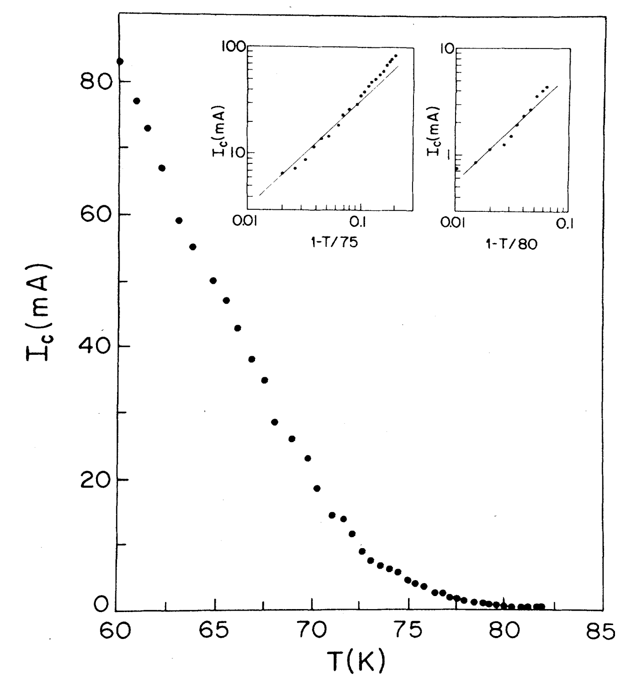

FIG. 2. Critical current vs temperature curve of sample with nominal composition $Y_{1.2}Ba_{0.8}Cu_2O_x$. This sample has a low resistivity ratio, high room-temperature resistivity, and a coarse grain structure with inhomogeneous composition. The insets show the same data presented as logarithmic plots of I_c vs $1-T/T_c$ in two ranges of temperature. The solid lines have slope one.

In contrast to this behavior Fig. 3 shows the temperature dependence of the critical current for a number of good-quality Y and Dy samples. Thus, all of them had low room-temperature resistivities and resistivity ratios around two. The data are shown in the form of a logarithmic plot of I_c vs $1-T/T_c$ where I_c is defined as the value of the current producing a voltage of 0.5 μV to 5 μV (depending on the sample and on the precision of the instruments) and T_c is the value of the temperature near the transition that produces the best fit of the data to a power law of the type $I_c \propto (1-T/T_c)^{\nu}$. Except for a very small region very close to T_c, the variation of I_c with temperature is clearly linear, that is $\nu=1$, down to the lowest temperatures measured where the critical current reached values near 0.5 A. This is true for both types of samples that we have fabricated, those with Dy and those with Y. A linear dependence of the critical current near T_c is characteristic of Josephson-coupled weak links. Even if the whole sample is not a superconductor, a percolative path exists between the two extremes of the sample. If this path is formed by grains in close proximity or separated by insulators, the dominant intergrain coupling is Josephson-like which would yield the appropriate temperature dependence of the critical current. Note that it is the weakest of such links in the system which determines the measured critical current.

To understand in more detail the temperature dependence of all our samples, we need to consider their morphology. They are composed of grains of typical size a_0 with varying composition and which may be Josephson coupled to each other. Neglecting the possible anisotropy in the critical current in these crystals, which would represent only a refinement, the temperature dependence of the critical current of the sample is determined by a number of multiply connected percolating clusters. The coupling strength between grains should have a wide distribution since the morphology of the sample corresponds to a

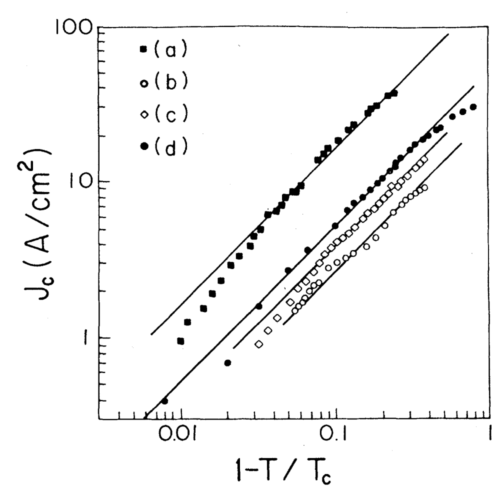

FIG. 3. Logarithmic plots of I_c vs $1-T/T_c$ curves of various samples. The linear dependence can be clearly seen in these plots. (a), (b), and (c) correspond to samples with nominal composition $YBa_2Cu_3O_y$ and (d) corresponds to a sample with composition $DyBa_2Cu_3O_y$. The solid lines have slope one.

three-dimensional continuum percolating structure.[9] Then, if the temperature dependence of the critical current of a pair of coupled grains varies as $F(T)$, the different magnitudes of I_c for each pair of grains will simply affect the magnitude of the sample critical current but its temperature dependence will still follow the functional form of $F(T)$, as long as the distribution of critical temperatures is sufficiently narrow so that most of the grains which are superconducting are already below their value of T_c.

Then, to understand our data what remains to be determined is the form of $F(T)$. For weak links in the dirty limit, $I_c(T)$ is determined by the Ambegaokar-Baratoff[10] relationship

$$I_c(T) = [\pi\Delta(T)/2eR_n]\tanh[\Delta(T)/2k_BT] , \qquad (1)$$

which near T_c reduces to $I_c(T) = \pi\Delta^2(T)/4k_BT_ceR_n$ and is proportional to $1-T/T_c$ in the Bardeen-Cooper-Schrieffer (BCS) theory. Since the new high-T_c materials are closer to the clean limit because they have such short coherence length and longer mean-free paths, the actual form is modified,[11] but the temperature dependence remains linear near T_c. On the other hand, independent of whether the links are tunnel junctions or continuous weak links in either the dirty or the clean limit,[11] $F(T)$ varies linearly with temperature near T_c.

Thus, in all cases, even though the sample is a three-dimensional array of Josephson junctions, the temperature dependence of the critical current will then be the same as the temperature dependence of the critical current of one Josephson junction. Such linear dependence was observed down to the lowest temperature T_m measured where the critical current reached the maximum current that our source can supply. For typical samples, T_m is between $0.9T_c$ and $0.7T_c$. This range is comparable to the range in which a linear dependence of the critical current has been measured[12] in single Josephson junctions of both tunneling and continuum type. Similarly, Eq. (1) is very close to a straight line down to temperatures as low as $0.7T_c$ as it can be seen in the figure presented by Ambegaokar and Baratoff.[10]

A simple way to understand the two different regimes is to consider two weakly coupled superconducting grains. If the current flows through them, then there are two characteristic critical currents: that of the individual grains I_{cg} which according to the Ginzburg-Landau theory varies as $(T_c-T)^{3/2}$ and that of the Josephson junction between them which is given by Eq. (1) and that near T_c varies as (T_c-T). Then, there will always be a temperature T_x, no matter how close to T_c, above which the $(T_c-T)^{3/2}$ behavior dominates. Clem *et al.*[7] have calculated the details by considering the relationship between the condensation energy of the typical grains in the sample and the characteristic Josephson coupling energy between them.

To consider these effects Clem *et al.*[7] compare the ratio $\epsilon = E_j/2E_c$ where $E_j(T) = \hbar I_c(T)/2e$ is the Josephson coupling energy between two grains and $E_c(T) = [H_c(T)^2/8\pi]V$ is the condensation energy. V is the volume of a grain and H_c is the thermodynamical critical field. If $\epsilon \ll 1$ the temperature dependence is linear and in the opposite limit it follows a $(1-T/T_c)^{3/2}$ dependence. Thus, we need to estimate ϵ to determine the temperature dependence of our samples. Following Clem *et al.*,[7] $\epsilon(T) = \epsilon_0 G(T)$, where $G(T)$ is a temperature-dependent function and where

$$\epsilon_0 = 2.93\hbar k_B/e^2\gamma T_c\rho_n a_0^2 . \qquad (2)$$

$G(T)$ diverges at $T=T_c$ so that any system will cross over from the linear regime to the $\frac{3}{2}$ regime sufficiently close to T_c. In much simpler terms, the crossover temperature T_x from one behavior to the other is given by

$$T_x = T_c(1-0.882\epsilon_0) , \qquad (3)$$

which gives a width of $T_c - T_x = 0.8$ K for $\epsilon_0 = 0.01$ and $T_c = 90$ K. Using estimated and measured parameters for the Y-Ba-Cu-O system [the Sommerfeld constant $\gamma = 3-5$ mJ (mol Cu)$^{-1}$ K^{-2} (Ref. 13), the measured resistivity of $\rho_n = 1000\ \mu\Omega$ cm the critical temperature $T_c = 90$ K, and the mass density of $d = 5.6$ g/cm^3], then Eq. (2) gives ϵ_0 between 2×10^{-2} and 2×10^{-6} for grain size between 0.01 and 1μm, respectively.

Experimental measurements[14] of the specific heat in the RBa$_2$Cu$_3$O$_7$ family of superconductors gives similar results for R=Y, Dy, Eu, Gd, Er. From that result we can infer that γ has the same value in all our samples. On the other hand, the mass density d, ρ_n, and T_c measured in our Dy-Ba-Cu-O samples are very close to those measured in the Y-Ba-Cu-O samples. Consequently, ϵ_0 has similar magnitudes for the same range of grain size. From the I_c vs $1-T/T_c$ curves we have obtained $1-T_x/T_c$ and found it to be between 0.06 and 0.006 which corresponds to ϵ_0 between 0.07 and 0.007. Equation (2) is satisfied if the effective grain size a_0 is between 0.005 and 0.02 μm. These values are much less than the size of the grains as observed in the microscope but correspond to the characteristic size of the intragrain domains between twin boundaries which thus determine the scale of the percolating network of Josephson junctions. Note that these ceramics have much lower values of ϵ_0 than other materials such as NbN or Al.

An apparent major weakness of the argument presented above is that Eq. (2) contains parameters from the BCS theory, the details of which do not necessarily apply to high-T_c oxides. In fact, as long as the Josephson effects are also present in these materials,[15] the Ginzburg-Landau theory should apply away from the fluctuation regime.[16] Then, $\epsilon(T)$ is simply a measure of the ratio of the coherence length to the grain size. Since estimates place the coherence length in the range of 1–4 nm,[17] then our samples should clearly be in the regime of linear dependence. Unfortunately, we have not been able to determine with certainty the temperature dependence of I_c for $T_c > T > T_x$ due to its small magnitude in that range of temperature. Therefore, we cannot conclude whether I_c obeys the Ginzburg-Landau theory[7] or if, on the contrary, fluctuations effects are dominant in this range of temperature as suggested by Lobb[16] and, consequently, I_c departs from the $(1-T/T_c)^{3/2}$ dependence.

Figure 4 shows a set of current-voltage characteristics of one of the YBa$_2$Cu$_3$O$_y$ samples. However, the shape of

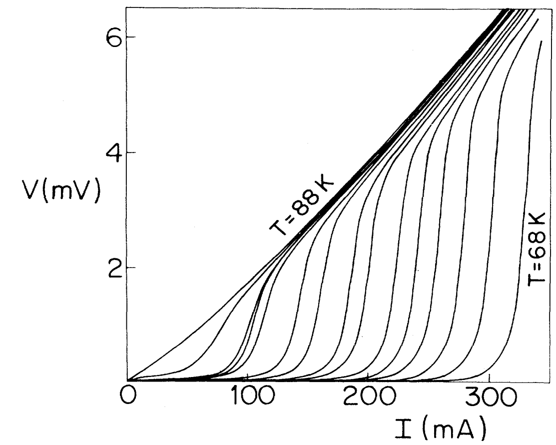

FIG. 4. Current-voltage characteristics of one of the $YBa_2Cu_3O_y$ samples. In order to determine the critical current, the *I-V* curves are measured with a voltage scale two hundred times smaller than that shown in this figure.

the *I-V* curves of a three-dimensional percolating network of Josephson junctions is still an open question from both theoretical and experimental points of view and it is certainly not the same as the *I-V* curve of an isolated Josephson junction. Consequently, no conclusion was derived from the study of the shape of the *I-V* curves of our samples.

Tinkham[8] proposes that, if the critical current is limited by flux creep then, I_c can vary as $1-T/T_c$ very close to T_c and under certain conditions, namely, that the parameter

$$\alpha \approx [k_B T_c / F_p(0)] \ln(v_0 / v_{\min}) \approx 1,$$

where F_p is the pinning energy and $v_{\min}$ is the minimum creep velocity detectable. This condition is by far more stringent than others discussed in the present paper.

It is worth while to point out that changing T_c by ± 1 K does not affect the exponent ν in our experimental I_c vs $(1-T/T_c)$ curves, being always 1 in the range of temperature $T<T_x$. However, for $T_x<T<T_c$, changing T_c by 0.5 K or less, strongly modifies the magnitude of the exponent ν, attaining values between 1 and 3 in that range of temperature. We are at the present carrying more sensitive measurements of I_c very close to the transition and at voltage levels smaller than 1 μV.

Finally, we comment on the prediction of Deutscher and Müller[18] of a critical current temperature dependence of the form $(T_c-T)^2$. Clearly, this type of behavior is not observed in any of our samples except for its possible existence very close to T_c. This prediction is based on the fact that the boundary condition for the pair potential can be written in general as

$$(\psi^{-1} d\psi/dx)_{x=0} = b^{-1}, \tag{4}$$

where b is an extrapolation length which describes the penetration of the pairs into the normal or insulating region.

We should first note the fact that such a temperature dependence of the critical current has never been observed even in superconductor–normal-metal–superconductor (SNS) junctions made with conventional superconductors where such dependence is also predicted. A more serious limitation is however, the fact that, except in a very narrow regime near T_c, in the high-T_c superconductors the Ginzburg-Landau coherence length $\xi(T)$ is of the order of the Pippard coherence length ξ_0 both of which are comparable to the interatomic distance a. Then, $\xi_0 \sim a \sim \xi(T)$ and thus the extrapolation length $b \sim a \sim \xi(T)$, so that we are in the regime $b/\xi(T) \sim 1$. On the other hand, since these materials are in the clean limit, $\xi(T)=0.74\xi_0/(1-T/T_c)^{1/2}$, and consequently, only within a few millikelvins near T_c, the depression of the gap will be significant enough to modify the temperature dependence of the critical current.

In summary, we have measured the temperature dependence of the critical currents in bulk samples of Y-Ba-Cu-O and Dy-Ba-Cu-O and we have found that I_c varies linearly with (T_c-T) from the lowest temperature measured to a certain temperature T_x close to T_c. The magnitude of T_x-T_c agrees with theoretical models of granular superconductors if the effective grain size a_0 is between 0.005 and 0.02 μm. These values are much less than the size of the grains as observed in the microscope. We conclude that intragrain domains between twin boundaries determine the scale of the percolating network of Josephson junctions.

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