Non-Ohmic Dissipative Regime in the Superconducting Transition of Polycrystalline $Y_1Ba_2Cu_3O_x$

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We have measured the low-field magnetoresistance and current-voltage characteristics of polycrystalline $Y_1Ba_2Cu_3O_{7-\delta}$ near the superconducting-normal transition. The development of a fully superconducting state occurs via an intermediate phase about 2 K in width. Within this intermediate phase, sample dissipation is highly non-Ohmic and exhibits extreme sensitivity to small magnetic fields. The nonlinear *I-V* characteristics describe a new phase with zero resistance and zero critical current which seems to arise from the presence of strong junction disorder.

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We report detailed measurements of the magnetoresistance and current-voltage characteristics of polycrystalline $Y_1Ba_2Cu_3O_{7-\delta}$ in a narrow temperature interval near the superconducting transition. These measurements, which support the view that polycrystalline $Y_1Ba_2Cu_3O_{7-\delta}$ is a weakly coupled granular superconductor,[1,2] suggest that the onset of superconductivity in this material occurs via a dynamical process whose behavior is substantially more complex than previously believed.

In zero magnetic field, we find that the development of a fully superconducting state occurs via an intermediate phase, bounded from above by a zero-resistance temperature, T_{c1}, and from below by a "zero dissipation" temperature, denoted by T_{c2}. Above T_{c1} the material is Ohmic and exhibits the resistive transition of a conventional fluctuation-broadened superconductor.[3] This broadened resistive transition is associated with the onset of superconductivity in individual grains; the single-grain transition temperature is customarily referred to as T_{c0}.

Immediately below T_{c1}, the electrical resistance of the superconductor appears to be zero, in the sense that $dV/dI=0$ in the limit of small measuring current. However, the superconductor does not possess a critical current in the intermediate regime, behaving instead as a non-Ohmic resistor with nonlinear current-voltage (*I-V*) characteristics. The *I-V* characteristics have a strongly temperature-dependent power-law form given by $V \sim I^{a(T)}$, with the exponent $a(T)$ increasing from unity (Ohmic behavior) as the temperature is lowered below T_{c1} and ultimately diverging at T_{c2}. Below T_{c2} the material behaves as a conventional superconductor, whose nonzero critical current is inferred by induced diamagnetic screening currents and the existence of a Meissner effect.

Our second principal finding is that the nature of the transition in the intermediate regime is strongly influenced by a weak external magnetic field. In an external field of the order of 1 G, the *I-V* characteristics (not shown here) in the intermediate regime become Ohmic at low current levels, in essence reducing T_{c1}. At higher current levels, the *I-V* characteristics retain their $B=0$ nonlinear character, raising the possibility that the magnetic field is superimposing an Ohmic response onto the non-Ohmic zero-field behavior. As discussed below, this sensitivity to small fields is a consequence of the weak coupling between grains. A more extensive analysis of the magnetic field results will be given in a separate publication.

Polycrystalline samples of $Y_1Ba_2Cu_3O_{7-\delta}$ were prepared by the solid-state reaction technique,[4] with the final pressed pellet sintered in air at 900 °C for 12 h and annealed in flowing O_2 at 500 °C for 12 h. Scanning electron microscopy examination shows a spread in grain diameters of 5–20 μm with an average of about 10 μm. Density measurements indicate that the samples are 30% porous. X-ray analysis shows only the 1-2-3 phase and places an upper bound of 3% on foreign phases. The room-temperature resistivity of the samples, whose typical dimensions are $1.0\times0.7\times0.2$ cm^3, is $\simeq 2.0$ mΩ cm at room temperature and falls linearly to $\simeq 0.8$ mΩ cm at 100 K.

Contact pads for lead attachment are prepared by ion milling of the sample and then sputtering of gold pads through a mechanical mask, resulting in a contact resistance of less than 0.1 Ω. Sample voltage is measured with a four-terminal, low-frequency lock-in technique with a noise floor of 1 nV. Sample temperature is measured with a Pt resistance thermometer with an absolute accuracy of 0.1 K and a reproducibility of better than 0.1 mK. Temperature regulation holds the sample temperature constant to 1 mK during measurements. A solenoid around the sample stage produces fields up to 100 G with a homogeneity of better than 0.1%. For all measurements reported here, the field is applied perpendicular to the direction of sample current flow. The sample stage and magnet coil are mounted in a liquid-nitrogen cryostat surrounded by a Mumetal shield that reduces the ambient field to <0.05 G.

Figure 1 shows voltage vs temperature for sample A at a fixed current of 1 mA, while Fig. 2 shows similar data for sample B (plotted logarithmically, to highlight the

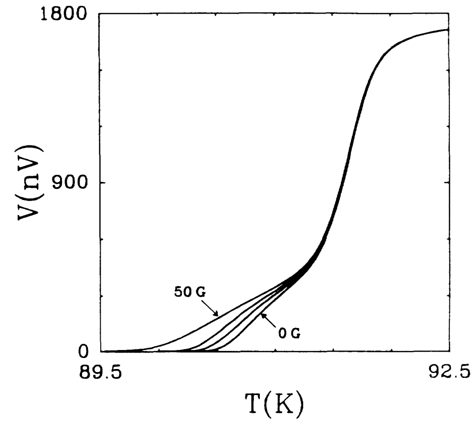

FIG. 1. Sample voltage vs temperature at constant current $I=1.00$ mA for sample A, $Y_1Ba_2Cu_3O_{7-\delta}$. Data were taken by field cooling of the sample in applied magnetic fields of 0, 2, 6, and 50 G.

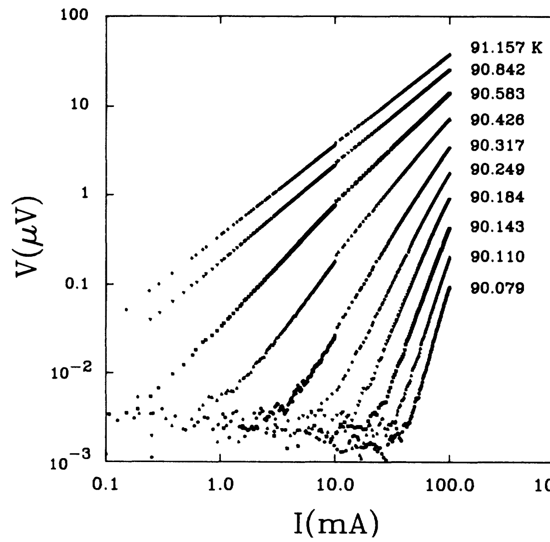

FIG. 3. Current-voltage characteristics for sample A in zero magnetic field at various temperatures in the "foot" of the transition region.

exponential behavior at low temperatures). Three other samples have been studied to date with similar results. The plots in Figs. 1 and 2 have the character of the "double transition" previously observed in weakly coupled granular superconductors.[5] At the bulk transition temperature T_{c0}, a sharp drop in the resistance signals the single-grain transition. At lower temperatures, where intergrain coupling energies exceed the thermal energy, phase ordering occurs and the sample enters a zero-dissipation state. In the intermediate temperature range, however, the sample exhibits an unexpected non-Ohmic behavior.

This unexpected behavior is illustrated in Fig. 3, where the *I-V* characteristics of sample A are plotted in this intermediate temperature range. Throughout our experimental window ($V > 1$ nV, $I < 100$ mA), the data exhibit simple power-law behavior, with $V \simeq I^{a(T)}$, and $a > 1$.

The temperature dependence of $a(T)$ is shown in Fig. 4 where, for comparison, the $B=0$ voltage data of Fig. 1 are also shown. Below T_{c1}, $a(T)$ increases from one, apparently diverging at a temperature T_{c2} of about 90 K. The solid line in Fig. 4 shows a fit by the form $a(T) = \exp[A(T_{c1}-T)/(T-T_{c2})]$, with $A=0.3$, $T_{c1}=91.3$ K, and $T_{c2}=89.8$ K. The apparent divergence of $A(T)$ suggests definition of T_{c2} as the temperature at which

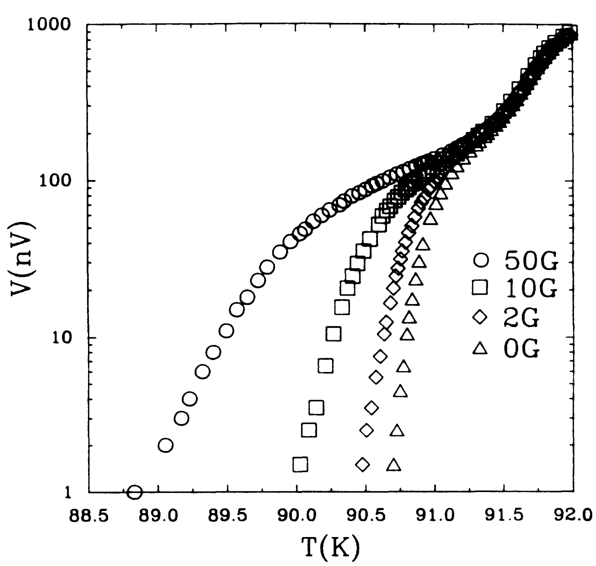

FIG. 2. Sample voltage vs temperature at constant current $I=1.00$ mA for sample B, $Y_1Ba_2Cu_3O_{7-\delta}$.

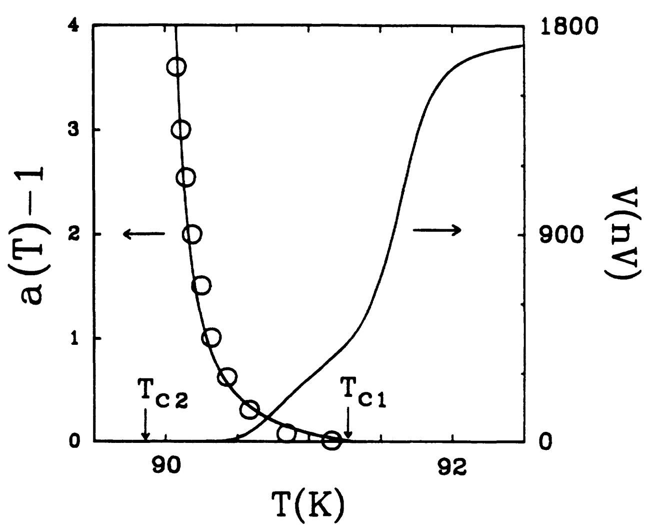

FIG. 4. *I-V* exponent $a(T)-1$ and zero-field voltage curve vs temperature. Note that the footlike structure in $V(T)$ is an artifact of the nonlinear current dependence. The line through the data points is a fit by an exponential divergence (see text) with $T_{c1}=91.3$ K and $T_{c2}=89.8$ K.

$a(T)$ diverges and below which the sample displays a critical current. We emphasize that this non-Ohmic behavior implies that the shape of the zero-field "foot" in Figs. 1 and 2 depends on the measuring current. The foot becomes less pronounced at lower currents.

As seen in Figs. 1 and 2, the sample dissipation in the intermediate temperature region is strongly influenced by a weak external magnetic field.[6] Unlike the zero-field data, which reflects non-Ohmic dissipation, the nonzero-field *I-V* characteristics (not shown here) are purely Ohmic at low current densities ($j < 0.04$ A/cm^2, corresponding to $I < 5$ mA). At higher current levels, there is a crossover to non-Ohmic power-law behavior, with an apparent divergence in $a(T)$ at lower temperatures. Thus in a magnetic field of 2 G or greater, the foot structure reflects a real sample resistance which, as is evident from Fig. 2, decays exponentially with decreasing temperature.

One other feature in the data not evident in Fig. 1 is that the steep clifflike portion of the curve just above T_{c1} is shifted down in temperature by a magnetic field. High-resolution measurements (not shown here) show a shift in temperature of 0.25 ± 0.02 mK/G. This shift is consistent with measurements of dH_{c2}/dT in single-crystal samples,[7] indicating that the "cliff" indeed corresponds to the single-grain transition.

Figure 5 is a plot of "T_c" versus field, where "T_c" is defined as the temperature at which the sample voltage falls below a level corresponding to a sample resistivity of 10^{-6} Ω cm (about 5 nV with a 1-mA sample current). At low fields (<2 G), "T_c" so defined is current dependent because of the non-Ohmic behavior. In this sample, at fields below 5, G d"T_c"$/dB$ is about -0.06 K/G, decreasing in magnitude to about -0.01 G/K at 50 G. These values vary from sample to sample by about 30%.

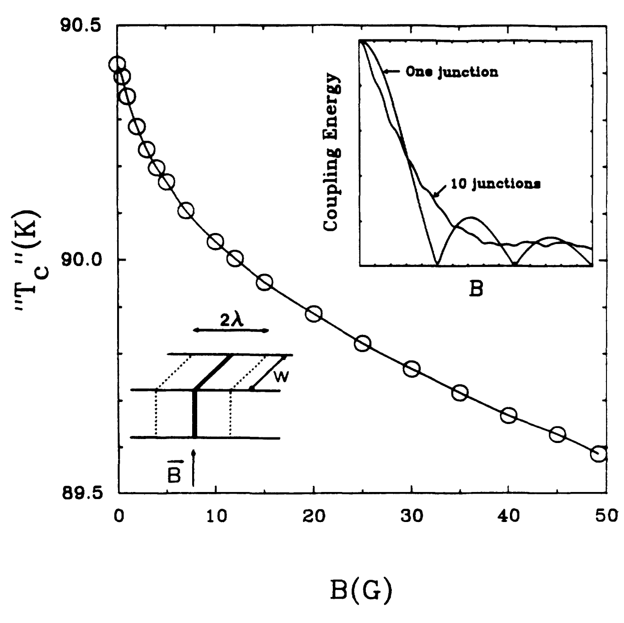

FIG. 5. "T_c" vs applied field, sample A. "T_c" is the temperature at which the sample voltage falls below 5 nV with a sample current of 1 mA.

The sensitivity to the first few gauss displayed in Figs. 1, 2, and 5 can be understood in terms of field penetration in the junctions between grains. The single-crystal penetration depth in this material is $\lambda(T) = \lambda_0[T_{c0}/2(T_{c0}-T)]^{1/2}$, where $\lambda_0 = 570$ Å is taken to be the geometric mean of λ measured parallel and perpendicular to the Cu-O planes.[7] Note that $\lambda(T_{c2}) \approx 4000$ Å, large but still much less than the 10-μm average grain size. Near T_{c0}, there is no strong phase coherence among the grains, so that material cannot screen out external fields. Even small fields freely penetrate the sample, threading the void space and the junctions between grains. When the field is parallel to a junction face (as shown in the lower inset to Fig. 5), it penetrates an area $2\lambda(T)w$, where w is the width of the junction. The critical current $i_c(T)$ and coupling energy $E_c(T) = \hbar i_c/2e$ of the junction are substantially reduced when the applied field is on the order of or larger than $\Phi_0/2\lambda(T)w$, where Φ_0, the flux quantum, is 2×10^{-7} G cm^2. If we take $w = 5$ μm and $\lambda(T_{c2}) = 4000$ Å, this field is about 5 G at temperatures near T_{c2}. The upper inset to Fig. 5 shows the coupling energy versus field for a single Josephson junction as well as the average coupling energy versus field for ten Josephson junctions with a ten-to-one spread in junction areas. (For a superconductor-normal-metal-superconductor junction, the dependence of coupling energy on field has a similar structure.) We expect the coupling energy and hence T_{c2} to be governed by such a curve, suitably averaged over junction sizes and orientations. The biggest reduction in T_{c2} will occur for fields up to $\Phi_0/2\lambda(T_{c2})w \approx 5$ G with a smaller effect at higher fields, precisely the behavior observed in Fig. 5.

These results reinforce the idea that the disorder in the intergrain coupling plays an important role in the non-Ohmic temperature region between T_{c1} and T_{c2}. Our samples possess a broad distribution of junction strengths, both because of the random orientations of the anisotropic grains and because of the distribution of junction areas. This disorder would cause the most strongly coupled grains to form tenuous percolating clusters of reduced effective dimensionality as the temperature is lowered below T_{c0}. In this context, it is useful to note that a fit to the data of Fig. 2 shows that at low temperatures $V \simeq \exp(-b/T)$, where b is a field-dependent constant. This exponential behavior is similar to that predicted and observed[8] in one-dimensional superconducting filaments, raising the possibility that the dissipative behavior is dominated by a quasi-one-dimensional percolating network of junctions.

The *I-V* characteristics of a two-dimensional array of junctions also exhibit power-law behavior.[9,10] In the Kosterlitz-Thouless picture for phase transitions in two dimensions, this power-law behavior arises from current-induced depairing of bound vortex-antivortex pairs. However, an extension of this picture to regular three-dimensional arrays (current-induced blowup of "flux tubes") yields exponential I-V characteristics, i.e., $V \simeq \exp(-a/Ik_BT)$, although this estimate neglects the effects of a distribution of junction strengths.

Although the mechanism for the nonlinear behavior we observe is not known, one expects the temperature region close to the transition to be fluctuation dominated and to exhibit non-Ohmic behavior. However, simple power-law behavior can be easily extracted from a homogeneous three-dimensional material. Its appearance here suggests that junction disorder reduces the effective dimensionality of the grains in polycrystalline $Y_1Ba_2Cu_3O_{7-\delta}$, and that the transition is actually a sequence of transitions first involving filamentary superconductivity (between T_{c1} and T_{c2}), followed by phase locking of the filaments below T_{c2} to form long-range three-dimensional order.

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[6]The data in Figs. 1 and 2 were obtained by field cooling of the sample. In measuring voltage vs field in the "foot," we find a complicated hysteretic behavior which we ascribe to flux trapping in the interiors of the grains when the field exceeds $H_{c1} \approx 5$ G at $T \approx T_{c0} - 1$ K.

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