# Pair-Breaking Effect of High Current Densities on the Superconducting Transition in $YBa_2Cu_3O_{7-\delta}$

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The equilibrium resistive transition of $YBa_2Cu_3O_{7-\delta}$ epitaxial films was measured at high pulsed current densities $J$ to promote homogeneous current flow and free flux flow. The superconducting transition temperature $T_c$ was then depressed as a function of $J$ in fixed fields $H$. The shifts in $T_c$, defined near midtransition, scaled following Ginzburg-Landau behavior: $\Delta T_c(H,J)/T_c(H,0)=[J/J_0]^{2/3}$, where $J_0$ is a field- and current-independent constant.

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The formation of the superconducting state is governed by a competition between four energies: condensation, magnetic-field expulsion, thermal, and kinetic. The order parameter, which describes the extent of condensation into the superconducting state, is reduced as $T$, $H$, and $J$ are increased. The boundary in $T$-$H$-$J$ space that separates the superconducting and normal states is where the three parameters attain their critical values $T_c(H,J)$, $H_{c2}(T,J)$, and $J_d(T,H)$, and any one of these functions completely defines the boundary. So far, to our knowledge, the shift in the $T_c(H,J)$ boundary caused by the pair-breaking (depairing) action of high currents has not yet been demonstrated in any high-$T_c$ superconductor and only the locus $T_c(H,J\approx 0)$ has been measured. The commonly measured $J_c(H,T)$ boundary that separates $\rho=0$ from $\rho\neq 0$ behavior is a sample-microstructure-dependent quantity of lesser fundamental importance, although of pivotal importance to applications.

A related but separate motivation for measuring $\rho(T)$ at high $J$ is to reveal more fully the bulk-equilibrium behavior. The resistive transition shape is altered at low $J$ in two ways. For a finite $T_c$ distribution, a weak current is more likely to sample higher $T_c$ percolative paths rather than flow uniformly through the entire specimen cross section. Also the tail of the transition is affected by nonequilibrium processes such as flux pinning and flux creep which tend to lower the resistivity below the free-flux-flow value. Both effects tend to make the transition sharper and give an apparently higher $T_c$. High current densities tend to promote uniform current flow as well as free flux flow [1], resulting in a resistive transition that is broader but more intrinsic and representative of the entire sample volume. In addition to these extrinsic effects, high current densities may also alter the transition in various intrinsic ways: e.g., by modifying the nature of fluctuations. Presently a complete theoretical model for $\rho(T)$ does not exist.

The measurement of $J_d(T,H)$ [or equivalently $T_c(H,J)$] is made difficult in high-$T_c$ superconductors because of the large normal-state resistivity $\rho_n$, combined with the large $J_d(0,0)$—a consequence of the high $T_c$. Thus to produce a sizable shift $\Delta T_c(H,J)=T_c(H,J)-T_c(H,0)$ (distinguishable from flux pinning and inhomogeneity effects) involves high current densities that, in combination with the large $\rho_n$, can cause serious heating problems. In a typical continuous-dc (CDC) measurement, the temperature rise $\Delta T_h$ is larger than the intrinsic $\Delta T_c(H,J)$ by an order of magnitude. Further, the thermal-conduction problem involved in the calculation of $\Delta T_h$ is relatively complicated because of the long time scales.

In a previous Letter [1] we described an apparatus that allows precision pulsed-current (PC) measurements on microsecond time scales, resulting in negligible temperature shifts, at enormous dissipation levels. In this work, we have refined the apparatus to permit similar high-current measurements over the entire resistive transition—well into the normal state. Compared with a CDC measurement, the use of current pulses of short duration and low duty cycle greatly reduces the temperature shift (by a factor of $\sim 50$) for a given power dissipation density $p=P/V=J^2\rho$. Additionally the shorter time scales simplify the thermal-conduction problem allowing precise calculation of the small temperature shift. Rather than being concerned with merely minimizing heating to negligible levels, we carefully calibrate the total thermal resistance $R_{\rm th}$ between the film and the heat sink. In this way we can subtract the heating-related temperature shift $\Delta T_h=R_{\rm th}p$ [which is much smaller than the intrinsic $\Delta T_c(H,J)$ anyway] to deduce the true temperature. In this way we find the intrinsic shift $\Delta T_c(H,J)$, defined near midtransition, to fit the Ginzburg-Landau expression for the depairing effect [2,3]:

$$\Delta T_c(H,J)/T_c(H,0)=[J/J_0]^{2/3}. \quad (1)$$

The depairing current density at $T=0$ and $H=0$ is given by

$$J_d(0,0)=0.41J_0=\frac{cH_c(0)}{3\sqrt{6}\pi\lambda_{ab}(0)}=\frac{c\Phi_0}{12\sqrt{3}\pi^2\lambda_{ab}^2(0)\xi_{ab}(0)}.$$

The sample was a $c$-axis-oriented epitaxial film of



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$YBa_2Cu_3O_{7-\delta}$ on a (100) $LaAlO_3$ substrate. The film was deposited and postannealed by means of the $BaF_2$ process, described elsewhere [4]. The precise stoichiometry and postannealing conditions were chosen to produce films that were relatively defect-free and had single-crystal-like quality [1,5]. The patterned bridge that was measured had dimensions ($t \times l \times w$): 100 nm × 3 mm × 100 $\mu$m.

The sample resistance was measured by applying rectangular current pulses of width 6 $\mu$s and rise time 0.4 $\mu$s. To within 1%, the current was both constant over the duration and independent of sample resistance. The repetition frequency was 2.5 Hz and could be increased by a factor of 100 without causing noticeable cumulative heating. The sample voltage and current were measured 2.6 $\mu$s from the beginning of the pulse. Maximum systematic plus random error in $R$ (unrelated to heating) was $< 2\%$. More information about the apparatus can be found elsewhere [1,6].

Conduction of heat from the sample undergoes the following processes [7,8]: Thermal diffusion within the sample occurs essentially instantaneously; on the time scale of nanoseconds, phonons transfer heat across the interface between the film and substrate; heat diffuses within the substrate in microseconds and finally into the heat sink in milliseconds. For our time scale of $\tau = 2.6$ $\mu$s, heat flow will cause a temperature variation within the film $\Delta T_1(z) \simeq (p/\kappa_1)(tz - z^2/2)$, a temperature drop across the interface $\Delta T_2 = ptR_{bd}$, and a temperature drop within the substrate given by [9] $\Delta T_3(\tau) = 2.26pt\tau w/2(D\tau)^{1/2} \times [4(D\tau)^{1/2} + w]c_p$. The symbols are defined as follows: $z$ is the distance of a point within the film from the interface, $\kappa_1 \approx 0.02$ W/cm K is the thermal conductivity of $YBa_2Cu_3O_{7-\delta}$ [10], $R_{bd} = 1 \times 10^{-3}$ K cm$^2$/W is the thermal boundary resistance at the film-substrate interface [7,11], $\kappa_3 = 0.32$ W/cm K and $c_p \sim 1.3$ J/K cm$^3$ are the thermal conductivity [7] and specific heat [12] of $LaAlO_3$, and $D = \kappa_3/c_p = 0.25$ cm$^2$/s is its diffusion constant. The temperature variation over the thickness of the film is small ($\sim 5\%$ of total), and the total temperature rise can then be taken to be $\Delta T_h = \Delta T_1(z = t/2) + \Delta T_2 + \Delta T_3(\tau = 2.6 \times 10^{-6})$. The corresponding total thermal resistance is $R_{th} = \Delta T_h/p = (1.9 + 10 + 21) \times 10^{-9} = 33$ nK cm$^3$/W. An extremely convenient property of $R_{th}$ is its relative temperature independence ($dR_{th}/dT \sim 0.1\%$/K) over the temperature range considered here (85–110 K)—allowing accurate correction of sample temperatures with a single value of $R_{th}$, which is independent of $T$, $H$, and $J$.

Figure 1 shows resistivity data measured at different PC current densities at $T = 100$ K, where fluctuation effects are small. The observed positive current dependence can then be wholly attributed to heating and the data can be made to fall onto the solid line [$\rho(T, H = 0, J \to 0)$, CDC measurement at $J = 5$ A/cm$^2$] if plotted against their actual corrected temperatures: $T = T_{sink} + pR_{th}$, where $T_{sink}$ is the temperature of the sample block. The corrected data conform to the "intrinsic" $\rho(T)$ line within their scatter, yielding a value of $R_{th} = 24 \pm 1$ nK cm$^3$/W from the fit. This measured value is consistent with the previously calculated value, given the uncertainty in the parameters. In fact the method described above can be used as a technique for determining $R_{bd}$, $D$, and $c_p$ by measuring at three different time scales and simultaneously solving the thermal-conduction equations. One advantage of this method is that the film does not require special patterning (such as required by the Swartz-Pohl method) other than the standard four-probe pattern.

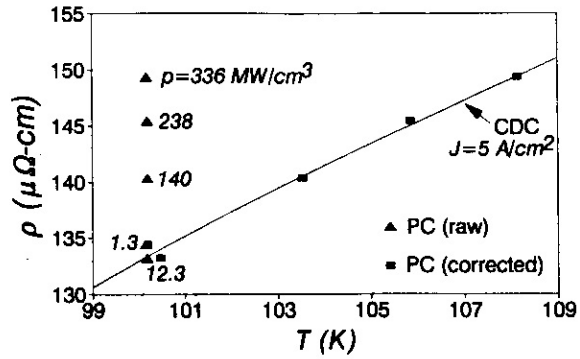


FIG. 1. The solid line is $\rho(T)$ measured with CDC. Symbols are PC data plotted against the raw (nominal) temperature $T_{sink}$, and the corrected temperature $T = T_{sink} + pR_{th}$.

Henceforth, all temperatures of data in the main experiment have been corrected with the $R_{th} = 24$ nK cm$^3$/W measured above. In order to maintain a 2% accuracy in the correction, the transitions were measured in fields no larger than 1 T to keep them sufficiently narrow.

Figure 2(a) shows a set of resistive transitions measured in $H = 0$ at different PC current densities. The success of the temperature-correction scheme is evident from the fact that the curves, at different $J$, all converge in the normal state (where intrinsic current dependence should disappear). In the transition region the curves are shifted downward in $T$ and become broader as a function of increasing $J$, as expected from the earlier discussion. Figure 2(b) shows similar data for $H = 0.3$ T. Also measured were data at $H = 1$ T [13]. Field orientation is $H \| c$. The shifts tend to become more parallel as $J$ is increased, because of progressive suppression of nonequilibrium effects as explained earlier. The shifts are most nearly parallel near $\rho = 40$ $\mu\Omega$ cm for all three fields. We thus assign the temperature at that resistivity level to $T_c(H, J)$. Figure 3(a) shows those $T_c(H, J)$'s, at different $H$ and $J$. Figure 3(b), which embodies the main result of this work, shows the scaled $T_c$ shift $\Delta T_c(H, J)/T_c(H, 0)$ plotted against $[J/J_0]^{2/3}$. The data at all $T$, $H$, and $J$ scale onto the line $y = x$ in agreement with Eq. (1). The adherence to the two-thirds power law is very close ($0.66 \pm 0.04$) and independent of any fitting

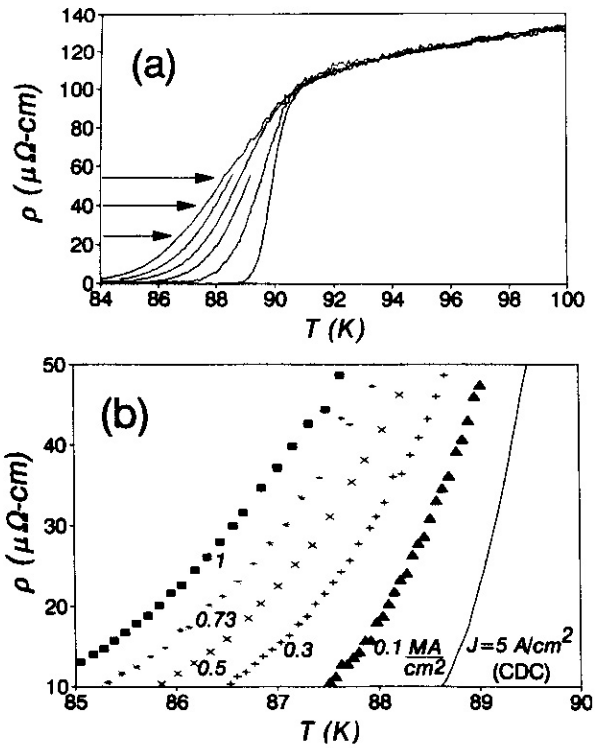


FIG. 2. $\rho(T)$ measured at different current densities [indicated in (b)] in fixed fields of (a) $H=0$ T and (b) $H=0.3$ T. Arrows in (a) show resistivity levels used in the analysis. In (a) only partial data are shown for intermediate currents, for the purpose of clarity.

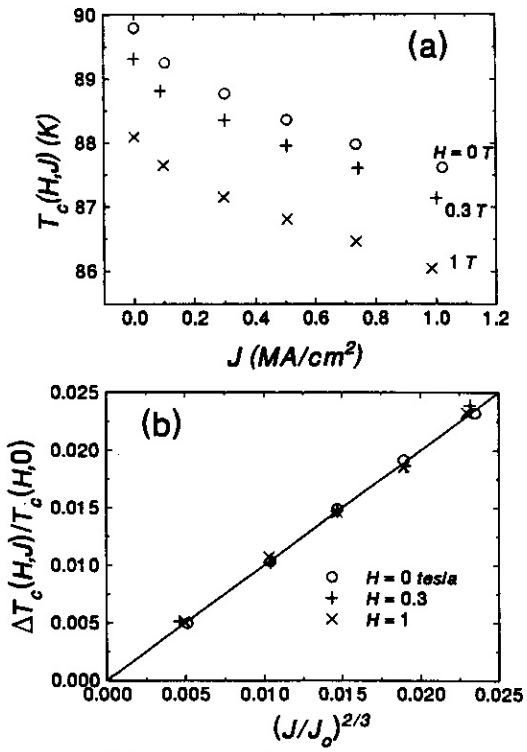


FIG. 3. Raw (a) and scaled (b) data showing dependence of $T_c(H,J)$ on $J$ at indicated fields. $J_0=2.9\times10^8$ A/cm$^2$ for all $H$. $T_c(H,0)\approx T_c(H)$ measured for $J\to 0$. Line is $y=x$.

parameter, further illustrating that the data are free from heating error; heating would raise the power-law exponent asymptotically to 2. All the data could be scaled with a single value of $J_d(0,0)=0.41J_0=1.2\times10^8$ A/cm$^2$, to bring the slope to unity. For each field, the parameter $T_c(H,0)$ was chosen to make the intercept zero. The resulting $T_c(H,0)$'s are close to $T_c'(H,0)$'s, the values measured using low-current (5 A/cm$^2$) CDC.

Recall that, in the above analysis, $T_c(H,J)$ was defined as the temperature where $\rho(T,H,J)=40$ $\mu\Omega$ cm. The analysis was repeated at two other resistivity levels: 25 and 55 $\mu\Omega$ cm. The qualitative behavior of Fig. 3(b) (i.e., the scaling and adherence to two-thirds power law) is preserved; however, the fits to unity slope gave $J_d(0,0)=0.83\times10^8$ and $1.7\times10^8$ A/cm$^2$ at 25 and 55 $\mu\Omega$ cm, respectively. Because of the observed broadening of the transition rather than a completely parallel shift, the $J_d(0,0)$ deduced in this way is sensitive to the resistivity-level criterion and should only be taken as a rough estimate. Note, however, that defining $T_c(H,J)$ near midtransition is not as arbitrary as it may seem. In an analogous experiment where resistive transitions broaden as functions of $H$, Jia *et al.* [14] have shown that the midpoint $T_c$ shift (for $H>1$ T) coincides with the shift in $T_c$ found by fitting to the complete fluctuation-flux-flow model [15]. In any case we find that our $J_d(0,0)=(1.2\pm0.4)\times10^8$ A/cm$^2$ is roughly comparable to the value $(3.3\times10^8$ A/cm$^2)$ calculated from currently known values [16] of $H_c$ and $\lambda_{ab}(0)$. Consistent with its role as an upper limit on the practical quantity $J_c$, $J_d(0,0)$ exceeds all measured values of $J_c$ [17].

Figure 4 compares values of $T_c(H,0)$ found from the intercepts of the scaling procedure against $T_c'(H,0)$, the actual values measured at very low current, and this was done for all three resistivity levels and fields. As can be seen, a slight departure between the two becomes noticeable only at the lowest resistivity level of 25 $\mu\Omega$ cm where

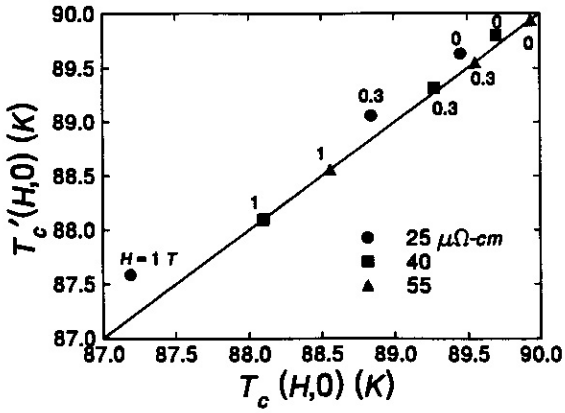


FIG. 4. $T_c(H,0)$ derived from the scaling [i.e., Eq. (1) or Fig. 3(b)] and $T_c'(H,0)$ measured for $J\to 0$. Line is $y=x$. Numbers indicate resistivity levels [see text and Fig. 2(b)] and fields.

flux motion departs from free flow at low $J$. The convergence of the temperatures at the higher $\rho$ indicates homogeneous current flow even at the low value of $J=5$ A/cm$^2$. This has a bearing on the observed anomalous sign reversal in the Hall effect [18]. There has been conjecture that the phenomenon could arise due to the extrinsic effect of percolating current paths [19]. The data in Fig. 4, however, tend to argue against such a mechanism.

In conclusion, the resistive transition was measured in a $YBa_2Cu_3O_{7-\delta}$ film at large pulsed-current densities to investigate the influence of pair breaking. The observed midtransition $T_c$ shift was found to follow the behavior predicted by Ginzburg-Landau theory [Eq. (1) and Fig. 3(b)]. It is hoped that these measurements will stimulate the development of a comprehensive theory of the resistive transition that considers pair breaking in the presence of fluctuations and flux motion.

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[1] M. N. Kunchur, D. K. Christen, and J. M. Phillips, Phys. Rev. Lett. **70**, 998 (1993).

[2] Michael Tinkham, *Introduction to Superconductivity* (McGraw Hill, New York, 1975); T. P. Orlando and K. A. Delin, *Foundations of Applied Superconductivity* (Addison-Wesley, Reading, MA, 1991). There is a weak magnetic-field dependence which has been neglected. It produces a $<1\%$ error for our largest applied field.

[3] Essentially the same expression can be obtained more generally in London theory simply by equating the kinetic-energy density to the condensation energy. This $J_d(0,0)$ is 1.84 times larger.

[4] M. P. Siegal *et al.*, J. Appl. Phys. **68**, 6353 (1990); D. J. Carlson *et al.*, J. Mater. Res. **5**, 2797 (1990).

[5] M. P. Siegal *et al.*, Appl. Phys. Lett. **60**, 2932 (1992).

[6] M. N. Kunchur, Bull. Am. Phys. Soc. **38**, 459 (1993).

[7] M. Nahum *et al.*, Appl. Phys. Lett. **59**, 2034 (1991), and references therein.

[8] We found that the temperature rise did not depend on whether the sample was in a vacuum or surrounded by helium vapor. Presumably the short time scales make convection ineffective. For this reason it also seems likely that direct contact with a liquid cryogen will not reduce heating significantly.

[9] S. K. Gupta *et al.*, Physica (Amsterdam) **206C**, 335 (1993), and references therein.

[10] S. J. Hagen, Z. Z. Wang, and N. P. Ong, Phys. Rev. B **40**, 9389 (1989).

[11] $R_{bd}$ is somewhat sample dependent because of its sensitivity to interface roughness.

[12] To the best of our knowledge, the specific heat of $LaAlO_3$ has not been published. The value taken is typical of other perovskite compounds such as $SrTiO_3$ and $YBa_2Cu_3O_{7-\delta}$; F. A. Hegmann and J. S. Preston, Appl. Phys. Lett. **62**, 1158 (1993); A. Schilling *et al.*, Physica (Amsterdam) **185-189C**, 1755 (1991); *Thermophysical Properties of Matter*, edited by Y. S. Touloukian (IFI/Plenum, New York, 1970), Vols. 2 and 5.

[13] In principle, the $H=0$ curves in Fig. 2(a) are at different self-fields ($\sim 10$ G at the highest $J=10^6$ A/cm$^2$). However, the resulting transition broadening was calculated to be negligible compared with the pair-breaking effect.

[14] Y. X. Jia *et al.*, Phys. Rev. B **45**, 10609 (1992).

[15] S. Ullah and A. T. Dorsey, Phys. Rev. B **44**, 262 (1991); Phys. Rev. Lett. **65**, 2066 (1990).

[16] J. G. Ossandon *et al.*, Phys. Rev. B **45**, 12534 (1992); U. Welp *et al.*, Phys. Rev. Lett. **62**, 1908 (1989); Z. Hao *et al.*, Phys. Rev. B **43**, 2844 (1991).

[17] There is one exception [H. Jiang *et al.*, Phys. Rev. Lett. **66**, 1785 (1991)] where the authors report $J_c=1.3\times10^9$ A/cm$^2$. However, close inspection of the information given implies an electric-field criterion 3–4 orders of magnitude greater than the accepted standard of 1 $\mu$V/cm.

[18] S. J. Hagen, C. J. Lobb, and R. L. Greene, Phys. Rev. B **43**, 6246 (1991), and references therein; J. P. Rice *et al.*, Phys. Rev. B **46**, 11050 (1992).

[19] J. Luo *et al.*, Phys. Rev. Lett. **68**, 690 (1992); E. C. Jones *et al.* (unpublished).