

Interlayer Tunneling Model for the c -Axis Resistivity in High-Temperature Superconductors

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The c -axis resistivity of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals shows a magnetic-field-dependent peak below the zero-field T_c . We find good agreement with the temperature and field dependence of the peak, by modeling the c -axis conduction as a series stack of Josephson tunnel junctions. For intermediate values of the Josephson coupling, we predict such a peak and verify it by measurements on discreet, thin-film Josephson junctions. The exaggerated effects in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ result from its unusually large energy gap and fluctuations.

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Knowledge of the c -axis transport in high- T_c superconductors (HTS) is essential for a complete understanding of the electronic states involved, and their c -axis coherency, which has implications for theory [1] and experiment [2,3]. Recent measurements of the c -axis resistivity ρ_c in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals show [4] a magnetic-field-dependent peak near the zero-field transition T_c for fields H parallel to the c axis, which has been confirmed by later studies [5]. Previous attempts to explain the field-dependent transition at low temperatures T by one-dimensional (1D) phase slippage [4] or by fluctuation effects [6] gave no explanation of the empirical normal-state "semiconducting" behavior.

We propose a *single* explanation for *both* the low- T , H -dependent transition and the high- T , H -independent normal-state semiconducting behavior by considering the c -axis conduction to be that of a series stack of Josephson tunnel junctions between well-coupled units consisting of superconducting Cu-O bilayers. Because of the large number of junctions along the c axis, each one is at its zero-bias resistance R_0 for practical bias voltages. As T decreases below T_c , tunneling theory predicts an increasing R_0 for Giaever junctions [7], with low values of the Josephson coupling parameter γ , while for large γ , R_0 decreases due to Josephson coupling [8–10]. For intermediate values of γ , we propose a model combining these effects, which displays a peak in R_0 below T_c , and verify it with data on discreet, thin-film junctions. We find that the relatively stronger semiconducting behavior of ρ_c in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$, extending to T well above T_c , requires, *in addition*, unusually large fluctuations [6,11,12] and energy gap Δ [13–15]. Using 2D fluctuations, calculated [12] beyond the Gaussian approximation, and Δ measured by tunneling [13–15], we find good agreement with the measured $\rho_c(T, H)$. As an *independent* check of this model, the fitting parameter γ predicts [8,9,16] a normal-state, c -axis resistivity, ρ_{cN} , which is close to the measured one.

Consider a single Josephson junction: the important parameter, γ , is the ratio of the Josephson coupling energy to $k_B T$, given by [10]

$$\gamma(T) \equiv \frac{\hbar I_c(T)}{ek_B T} = \frac{\pi \hbar \Delta(T)}{2e^2 R_N k_B T} \tanh \left\{ \frac{\Delta(T)}{2k_B T} \right\}, \quad (1)$$

where $I_c(T)$ is the Josephson critical current in the absence of fluctuations and R_N is the normal-state junction resistance. Note that both I_c and R_N are *extrinsic*, depending not only on the electron transmissivity but also the effective junction area. Here $\gamma_0 \equiv \hbar I_c(0)/ek_B T_c$ is appropriate for comparing junctions, and, for large R_N , $\gamma_0 \ll 1$, so Josephson coupling can be neglected at all T : then the junction resistance is solely due to quasiparticle tunneling and *for low bias voltages*, $V \ll \Delta$, it increases [7] with decreasing T as $\sim \exp[\Delta(T)/k_B T]$, since the quasiparticles are frozen out with the establishment of the gap. The quasiparticle conductance for two identical superconducting electrodes, $Y_{SS}(T)$, is given analytically in Taylor's thesis [17], and at T_c equals the normal-state conductance, $Y_N \equiv 1/R_N$. For $\gamma \gg 1$, the pair conductance shorts out the quasiparticle resistance, but true zero resistance only occurs asymptotically at $T=0$ as $\gamma(T)$ goes to infinity, since otherwise fluctuations in the phase difference across the junction lead to a finite voltage [10]. The conductance Y_{RSJ} for a resistively shunted junction is given by [10]

$$Y_{RSJ}(T) = Y_N I_0^2(\gamma(T)/2), \quad (2)$$

where I_0 is the modified Bessel function.

For the presently interesting case of $\gamma \sim 1$, we suggest that the total conductance of an *unshunted* junction is the sum of the quasiparticle and pair conductances, Y_{SS} and Y_p , the latter obtained by subtracting the shunt conductance, Y_N , from Eq. (2), i.e.,

$$Y_p(T) = Y_N \{ I_0^2(\gamma(T)/2) - 1 \}. \quad (3)$$

Thus as γ_0 increases, there is a smooth progression in the T dependence of R_0 from a purely decreasing function to a purely increasing function, but for $\gamma_0 \sim 1$, this model predicts a distinct peak in $Y_N = Y_{SS} + Y_p$ below T_c .

To verify this model, data on discreet Josephson junctions using two thin-film, Al electrodes are shown in Fig.

1 together with fits based on this model and a BCS dependence for $\Delta(T)$, which is verified to high precision in *these* junctions. Thus the only adjustable parameter is γ_0 , and these are shown for each curve. Note that the occurrence of this peak has also been recognized [18] in the transport resistance of Josephson-coupled granular superconductors.

From Eq. (1), one expects $\gamma_0 \propto 1/R_N$. However, as already noted even for small ($\sim 100 \mu\text{m}^2$) Nb Josephson junctions [19], the presence of film defects (e.g., grain boundaries) limits the Josephson correlated regions to areas, $A_0 \sim 1 \mu\text{m}^2$, which are generally smaller than the physical junction sizes, especially for the large ($\sim 1 \text{mm}^2$) Al junctions used here. Thus one uses $r_N A_0$ instead of the junction R_N in Eq. (1), where r_N is the specific junction resistivity. The Nb data agreed *quantitatively* with theory [8–10] using an effective junction area which interpolates between A_0 for $H=0$ and the high-field limit of Φ_0/H , according to $R_N = r_N/(A_0^{-1} + H/\Phi_0)$. This high-field limit of $\gamma(T)$, neglecting A_0 , was also used [16] to explain a crossover between 3D flux lines and 2D pancake vortices in highly anisotropic HTS and it equals the interlayer Josephson coupling energy for pancake vortices, offset in neighboring Cu-O bilayers [20] by a separation distance of the Abrikosov lattice spacing. At greater distances, this energy is truncated due to overlap with *other* vortices.

The use of this field dependence was already recognized [4] for the *c*-axis resistivity in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ below the peak, but a critical phase-slippage current, with a different T dependence than the above Josephson $I_c(T)$, was suggested as its origin. In addition, an *ad hoc* assumption [4] of an empirical H -independent “normal-state resistance” above the peak was needed.

In order to demonstrate that the field dependence of ρ_c has its origins in interplanar Josephson coupling, the effects of fluctuating superconducting regions above T_c must also be addressed, including finite H . The finite,

temporally and spatially averaged Ginzburg-Landau (GL) order parameter, $\psi \propto \Delta$, will remove quasiparticles above T_c , just as in the above case of a mean-field Δ for $T < T_c$, to reduce the quasiparticle conductance below Y_N . The magnitude of $\langle |\psi|^2 \rangle$ due to fluctuations has been calculated [12] after extending the GL theory beyond the Gaussian approximation and including finite H . For the extremely anisotropic $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$, we make the appropriate approximation of negligible interlayer coupling (i.e., an infinite effective mass ratio), so that the integral over electron momentum parallel to the field (and thus the *c* axis) is [12] replaced by $1/2d_c$, which reflects the localization of the electrons to a spatial dimension d_c , where d_c should be less than the repeat distance of the bilayers, s . Thus, we obtain

$$\langle |\psi|^2 \rangle = \frac{k_B T_c N_{2D}}{2d_c} t \sum_{n=0}^N (n + \tilde{\epsilon}_H/2h)^{-1}, \quad (4)$$

where $t \equiv T/T_c$, $N_{2D} \equiv m/2\pi\hbar^2$ is the free-electron 2D density of states (DOS), $h \equiv H/H_{c2}(0)$, $H_{c2}(0)$ is the linear extrapolation of the GL upper-critical field to zero temperature, $\tilde{\epsilon}_H$ is the renormalized $\epsilon_H = t + h - 1$ after the Hartree approximation [12], and N cuts off the summation over Landau levels if the magnetic length, $l_H = (\Phi_0/2\pi H)^{1/2}$, is smaller than the zero-temperature, in-plane coherence length $\xi_{ab}(0)$. Since $H_{c2} = \Phi_0/2\pi\xi_{ab}^2$, N is the largest integer smaller than $1/h$. Note that $\langle |\psi|^2 \rangle$ represents the magnitude of the order parameter in the *Cu-O bilayers*, of width d_c . The average over the unit cell is smaller, i.e., multiplied by a factor of d_c/s . To obtain the T -dependent $\langle |\psi|^2 \rangle$, the self-consistency condition [12] is needed:

$$t = \tilde{\epsilon}_H - h + 1 - \frac{k_B T_c}{8E_c(\pi\xi_{ab}^2 d_c)} t \sum_{n=0}^N (n + \tilde{\epsilon}_H/2h)^{-1}, \quad (5)$$

where E_c is the condensation energy per unit volume, $H_{c2}^2/8\pi$, and we have assumed that the GL parameter $\kappa \gg 1$. After summing over Landau levels, $\langle |\psi|^2 \rangle$ is found to be virtually field independent above T_c . There is one parameter left for $\langle |\psi|^2 \rangle$, and if values of H_{c2} and H_c are known, it is d_c . There is some semantical ambiguity here, since H_{c2} and H_c do not follow the $1-t$ dependence of the GL at low t , but saturate (e.g., in the clean limit, to one-half the extrapolated value). For $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$, appropriate values for $\mu_0 H_{c2}(0)$ and $\mu_0 H_c(0)$ are thus twice the measured values of 44 and 0.8 T, respectively, obtained from muon-spin-resonance experiments [21] and fits to the irreversibility line [22].

In order to fit ρ_c with the Josephson junction model, Δ is required at all $T < T_c$. We exploit the proportionality of $\langle |\psi|^2 \rangle$ in the GL theory to $\Delta(T)^2$ of the BCS theory near T_c , as shown by Gor'kov, to match the slope of the low- T extrapolation of $\langle |\psi|^2 \rangle$ from Eq. (4) to that of BCS near T_c . Using a straight-line tangent to both, we interpolate $\Delta_{in}/\Delta(0)$ at all T . Note that for finite H , both $T_c(H)$ and $\Delta(0)^2$ are reduced by a factor of $1-h$.

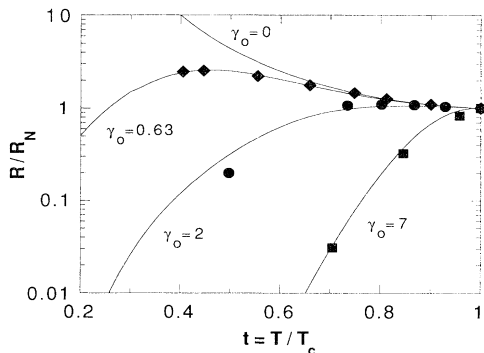


FIG. 1. Zero-bias resistance for discreet Josephson junctions using two thin-film, Al electrodes: diamonds, SG 3; circles, XIII; and squares, SG 9. The lines are fits, explained in the text, for the indicated values of γ_0 .

We use $\Delta_{\text{int}}/\Delta(0)$ and the zero-field $\Delta(0)$ from low- T tunneling [13–15] in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ to calculate the pair conductance from Eqs. (1) and (3), but the effective junction area for R_N in Eq. (1) must be considered carefully. By including the quartic term in the GL free energy, the fluctuation calculation [12] in a finite field shows that the coherence length $\xi(T)$ does not diverge at T_c , but that the coherence area is given by $\pi\xi(T)^2 = \pi\xi_{ab}(0)^2/\bar{\epsilon}_H$. At low temperatures in a magnetic field, however, we truncate this coherence area by the size of the lowest Landau-level orbit [11], i.e., Φ_0/H (we assume that A_0 is much larger).

In the zero-field ρ_c of Ref. [4], Josephson coupling only occurs for $T < T_c$, presumably because above T_c the superconducting regions are rapidly fluctuating and uncorrelated. Thus for the quasiparticle conductance above T_c we use that of a normal metal to superconductor junction, $Y_{NS}(T)$, for which an analytical solution is given on p. 45 of Ref. [17]. Although the quasiparticle conductance will shift towards $Y_{SS}(T)$ as T drops below T_c , we have ignored that in our calculations as it is difficult to quantify and, in any case, $Y_{NS}(T)$ is close to $Y_{SS}(T)$, i.e., $\sim 70\%$ – 80% . In fact, these calculated Y_{NS} and Y_{SS} assume as BCS DOS and equilibrium distribution functions, each of which may be only an approximation of the actual situation that may be better dealt with theoretically in a diagrammatic approach above T_c .

In Fig. 2, the data of Fig. 1 of Ref. [4] are shown as open symbols with a two-parameter fit with the above model (γ_0 to match the field-dependent resistance increase below the peak and d_c for $\langle|\psi|^2\rangle$ to match the field-independent shape above the peak). The present model thus explains *both the increase and decrease* of ρ_c with reasonable parameters: e.g., from the field-inde-

pendent decrease at high temperatures, $d_c \sim 0.6$ nm which is less than s , but greater than the Cu-O bilayer spacing of 0.3 nm implying that the bilayers are well coupled; while the field-dependent increase at low T gives $\gamma_0 \sim 4.1$ at 0.5 T and ~ 1.1 at 7 T, implying $\rho_{cN} \sim 1.4$ and 0.4 Ω cm, respectively. These values show internal consistency with the measured ρ_c , extrapolated to higher T , which is ~ 1 Ω cm (Fig. 1 of Ref. [4]). Note that the value of d_c , but not the quality of the high- T fit, is proportional to our choice for $H_{c2}(0)/H_c(0)^2$. Also shown in Fig. 2, as solid diamonds, are zero-bias tunneling resistance measurements in discreet break junctions [15] of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals. The excellent agreement with the measured ρ_c of Ref. [4] *above* the peak supports our contention that the origin of $\rho_c(H, T)$ is ordinary tunneling. Josephson coupling is weak in these high- R_N break junctions and is only seen at lower T (with $R_0/R_N \geq 10$).

Although γ_0 does not precisely follow the expected $1/H$ dependence, including A_0 can improve this. Also, however, the quality of the fit at 7 T is noticeably poorer than 0.5 T: Near the peak, the discrepancy is likely due to our neglect of $Y_{SS}(T)$ in the calculation, and that is consistent with the same deviations seen with the break-junction data in Fig. 2. However, at 7 T we do not understand the more abrupt onset of the experimental ρ_c at lower T .

The above fit predicts surprisingly large field-independent fluctuations of $\langle\Delta^2\rangle \sim 0.2\Delta(0)^2$ at T_c , so it is imperative to demonstrate consistency with other available data. A significantly smaller magnitude could not fit the experimental ρ_{cN} data for $T > T_c$. The fluctuation conductivity in the a - b planes, σ' , can be written [23] as a sum over electron momentum, k , as

$$\sigma' = \frac{2e^2}{m^*} \sum_k \langle|\psi_k|^2\rangle \tau_k, \quad (6)$$

where $m^* = 2m$, $\tau_k = \tau_0/(1 + k^2\xi_{ab}^2)$, and $\tau_0 = \pi\hbar/8k_B T_c \times (t-1)$. If we presume a predominant size for fluctuations to be ξ_{ab} , and thus approximate k by ξ_{ab}^{-1} , Eq. (6) becomes

$$\sigma' = \frac{e^2\tau_0}{2m} \langle|\psi|^2\rangle. \quad (7)$$

For comparison with experiment, we need the average conductivity over the unit cell, i.e., $\langle\sigma'\rangle = (d_c/s)\sigma'$. Evaluating $\langle|\psi|^2\rangle$ from the above fluctuation analysis [12] for $t=1.2$ gives $\langle\sigma'\rangle = 620$ mho/cm for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$. The best samples for comparison to this model, however, are low-defect, epitaxial films of $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$, shown in Fig. 3. We find a field independent [24] $\langle\sigma'\rangle = 550$ mho/cm at $t=1.2$, which agrees quite well with 650 mho/cm from the above calculation using the slightly smaller $s = 1.47$ nm for $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$. Another check comes from the agreement of the T -dependent break-junction tunneling conductances [15] shown in Fig. 2.

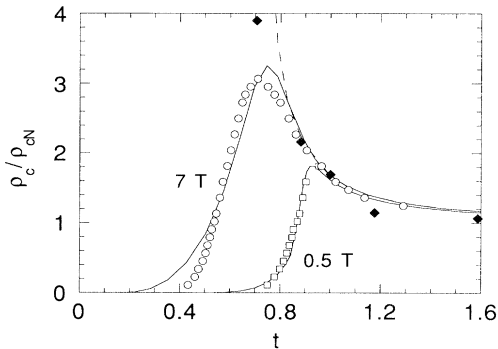


FIG. 2. Measurements of Ref. [4] for ρ_c of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystal (open squares, 0.5 T; open circles, 7 T) compared to our calculations for a series of Josephson junctions between Cu-O bilayers which includes the effects of fluctuations in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$, for the same two fields (solid lines) and with negligible Josephson coupling (dashed line). The latter is appropriate for the zero-bias tunneling resistance measurements in discreet break junctions [15] of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals (solid diamonds).

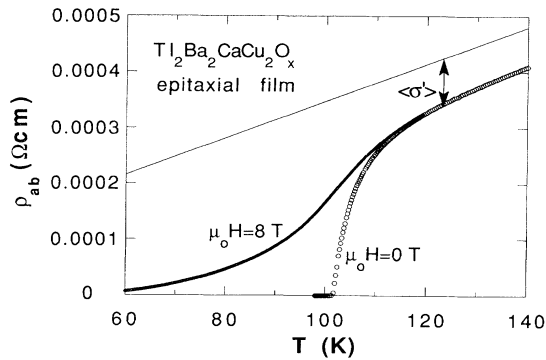


FIG. 3. The a - b plane resistivity measured in epitaxial films of $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ for fields of 0 and 8 T, parallel to the c axis. The line is a fit of a linear normal-state resistivity to this field-independent, high- T data using the temperature dependence for 2D fluctuations [see Eqs. (6) and (7)].

Our model explicitly includes Josephson coupling between Cu-O bilayers which is not contained in the Hamiltonian of Ref. [6]. The contribution of Ref. [6] was to include flux flow in the fluctuation analysis of the resistive transitions and for $\text{YBa}_2\text{Cu}_3\text{O}_7$ this gives excellent agreement with experiment: Apparently, because of the relatively strong coupling in this less-anisotropic material, the model works for ρ_c also. However, for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$, our analysis strongly suggests that phase fluctuations across the interplanar Josephson junctions dominate the dissipation.

In summary, we find that the unusual peak in ρ_c of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ can be quantitatively understood as a result of considering the material to be a series stack of ordinary Josephson tunnel junctions. Recently, a number of direct measurements [3] on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ crystals have also been shown to be consistent with such a model. It should be pointed out that severe compositional deviations or granularity can also cause increasing ρ_c at low T (i.e., semiconducting behavior), but the intrinsic tunnel-junction effect will always be present.

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- [1] P. W. Anderson, in *High-Temperature Superconductivity*, edited by J. Ashkenazi *et al.* (Plenum, New York, 1991), p. 1; R. A. Klemm and S. H. Liu, *Phys. Rev. B* **44**, 7526 (1991); N. Kumar and A. M. Jayannavar, *Phys.*

- Rev. B* **45**, 5001 (1992); A. J. Leggett (to be published).
 [2] K. Tamasaku, Y. Nakamura, and S. Uchida, *Phys. Rev. Lett.* **69**, 1455 (1992); S. L. Cooper, D. Reznik, P. Nyhus, M. V. Klein, W. C. Lee, D. M. Ginsberg, B. W. Veal, A. P. Paulikas, and B. Drabowski (to be published).
 [3] R. Kleiner, F. Steinmeyer, G. Kunkel, and P. Müller, *Phys. Rev. Lett.* **68**, 2394 (1992).
 [4] G. Briceno, M. F. Crommie, and A. Zettl, *Phys. Rev. Lett.* **66**, 2164 (1991).
 [5] Y. I. Latyshev and A. F. Volkov, *Physica (Amsterdam)* **182C**, 47 (1991); A. Kapitulnik, in *Proceedings of the Los Alamos Symposium, 1991* (to be published).
 [6] R. Ikeda, T. Ohmi, and T. Tsuneto, *Phys. Rev. Lett.* **67**, 3874 (1991); *J. Phys. Soc. Jpn.* **60**, 1051 (1991).
 [7] I. Giaever, H. R. Hart, and K. Megerle, *Phys. Rev.* **126**, 941 (1962).
 [8] P. W. Anderson, in *Proceedings of the Lectures at Ravello Spring School, 1963* (unpublished).
 [9] V. Ambegaokar and A. Baratoff, *Phys. Rev. Lett.* **10**, 486 (1963).
 [10] V. Ambegaokar and B. I. Halperin, *Phys. Rev. Lett.* **22**, 1364 (1969).
 [11] R. Ikeda, T. Ohmi, and T. Tsuneto, *J. Phys. Soc. Jpn.* **58**, 1377 (1989).
 [12] S. Ullah and A. T. Dorsey, *Phys. Rev. B* **44**, 262 (1991).
 [13] Q. Huang, J. F. Zasadzinski, K. E. Gray, J. Z. Liu, and H. Claus, *Phys. Rev. B* **40**, 9366 (1989); Q. Huang, J. F. Zasadzinski, K. E. Gray, E. D. Bukowski, and D. M. Ginsberg, *Physica (Amsterdam)* **161C**, 141 (1989).
 [14] J. X. Liu, J. C. Wan, A. M. Goldman, Y. C. Chang, and P. Z. Jiang, *Phys. Rev. Lett.* **67**, 2195 (1991).
 [15] D. Mandrus, L. Forro, D. Koller, and L. Mihaly, *Nature (London)* **351**, 460 (1991).
 [16] D. H. Kim, K. E. Gray, R. T. Kampwirth, J. C. Smith, D. S. Richeson, T. J. Marks, J. H. Kang, J. Talvacchio, and M. Eddy, *Physica (Amsterdam)* **177C**, 431 (1991).
 [17] B. N. Taylor, thesis, University of Pennsylvania, 1963 (unpublished), p. 50.
 [18] A. Gerber, T. Grenet, M. Cyrot, and J. Beille, *Phys. Rev. Lett.* **65**, 3201 (1990); C. Attanasio, L. Maritato, and R. Vaglio (private communication).
 [19] D. H. Kim, K. E. Gray, and J. H. Kang, *Phys. Rev. B* **45**, 7563 (1992).
 [20] J. R. Clem (private communication).
 [21] D. R. Harshman, R. N. Kleiman, M. Inui, G. P. Espinosa, D. B. Mitzi, A. Kapitulnik, T. Pfiz, and D. L. Williams, *Phys. Rev. Lett.* **67**, 3152 (1991).
 [22] When the irreversibility results are scaled to the thermodynamic measurements in $\text{YBa}_2\text{Cu}_3\text{O}_7$, a value of $H_c(0) = 0.8$ T is found for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$. See Fig. 3 of K. E. Gray, D. H. Kim, B. W. Veal, G. T. Siedler, T. F. Rosenbaum, and D. E. Farrell, *Phys. Rev. B* **45**, 10071 (1992).
 [23] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 251.
 [24] There is a small magnetoresistance which is not visible on the scale of Fig. 3. See D. H. Kim, K. E. Gray, R. T. Kampwirth, and D. M. McKay, *Phys. Rev. B* **43**, 2910 (1991).