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Quasi-two-dimensional phase fluctuations in bulk superconducting $YBa_2Cu_3O_7$ single crystals

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A detailed study of the electrical transport properties in high-quality *bulk* single crystals $YBa_2Cu_3O_7$ has manifested a Kosterlitz-Thouless (K-T) -type transition in the *ab* plane 0.2 K below the mean-field superconducting transition ($T_c^0=93.15$ K). The *c*-axis correlation length $l_c(T=0) \simeq 6$ Å and a large two-dimensional effective mass ($m_{ab}^*/m_e \sim 10$) of the supercarrier are estimated based on the K-T theory. The large effective mass suggests strong electron-electron correlations for the supercarriers confined in a quasi-two-dimensional sheet.

The large anisotropy in high-temperature superconducting oxides has been well-established experimentally,[1,2] and the anisotropic coherence lengths ($\xi_{ab} \gg \xi_c$) strongly suggest that the superconducting order parameter is much more correlated in the *ab* plane than in the *c* direction, and hence is indicative of quasi-two-dimensional behavior.[3,4] Although it is generally believed that the low dimensionality and strong electron-electron correlations are essential to the pairing mechanism of this class of superconductors,[5] direct experimental evidence of the quasi-two-dimensional nature has not been found. Furthermore, how the quasi-two-dimensional "sheets" couple together to give rise to the ultimate three-dimensional superconducting state is also a challenging issue. To gain more insight into these issues, accurate and detailed measurements on high-quality *bulk* samples are essential. In this work, we studied the electron transport properties of a high-quality single crystal in a narrow temperature interval [± 0.2 K around the mean-field (MF) superconducting transition temperature], where extrinsic flux pinning becomes insignificant, and intrinsic vortex behavior is manifested. The results show a Kosterlitz-Thouless[6] (K-T) -type behavior in the *ab* plane and a weak correlation along the *c* axis, hence the quasi-two-dimensional nature of the $YBa_2Cu_3O_7$ system in a bulk form is established. The implications of our experimental results on the superconducting mechanism, including a short *c*-axis correlation length ($l_c \simeq 6$ Å) and a large in-plane effective mass ($m_{ab}^* \sim 10 m_e$), are discussed.

In a two-dimensional superfluid or a superconducting thin film, vortices can be induced by thermal fluctuations, and are bound in pairs below the Kosterlitz-Thouless temperature ($T_{\text{K-T}}$) by the energy $U_v(T)=[q^2/\epsilon(T)] \times \ln[r/r_0(T)]$, where q denotes the corresponding topological charge of a vortex ($q^2=\pi n_s^{2D}\hbar^2/m^*=2k_BT_{\text{K-T}}$ for vortices of one unit charge, where n_s^{2D} and m^* are the two-dimensional density and the effective mass of the superfluid, respectively); r is the distance between two vortices, r_0 is the size of the vortex core,[6] and ϵ is a temperature-dependent dielectric constant which accounts for the screening and renormalization effects of the vortices. In general, an anisotropic superconducting system with an "easy-plane" anisotropy can manifest K-T behavior if the in-plane correlation length ξ_{ab} satisfies the condition $\xi_{ab} < \Lambda$,[7] where $\Lambda \equiv 2\lambda^2/l_c$, λ is the magnetic penetration depth, and l_c is the effective correlation length along the *c* axis. Since the correlation lengths ξ_{ab} and ξ_c in the new superconducting oxides are relatively shorter than those in the conventional superconductors, the K-T-type transition may be anticipated in a narrow temperature interval below the MF Ginzburg-Landau critical temperature T_c^0.

In the zero magnetic field limit, there are three signatures in the electrical transport properties for the K-T transition. The first one is associated with the onset of resistivity for temperatures slightly above $T_{\text{K-T}}$.[6,8,9] For $T_{\text{K-T}} < T \ll T_c^0$, vortices begin to unbind, and give rise to a finite resistivity.[8,9] The resistivity in this temperature range shows a temperature dependence

$$\frac{R}{R_N} \propto \xi_+(T)^{-2} = \xi_{ab}(T_{\text{K-T}})^{-2}\exp\left[-2b'\frac{(T_c^0-T_{\text{K-T}})^{1/2}}{(T-T_{\text{K-T}})^{1/2}}\right],$$

with ξ_+ given by the phase correlation length[8,9] of the order parameter, $\xi_{ab}(T)$ is the Ginzburg-Landau correlation length in the *ab* plane [$\xi_{ab}(T) \propto |1-t|^{-1/2}$, t is the reduced temperature, $t=T/T_c^0$], R_N is the normal-state resistance, and b' a nonuniversal constant of order unity. However, in the limit of $T_{\text{K-T}} < T \sim T_c^0$, other paraconductivity effects,[10,11] due to amplitude fluctuations, can interfere with the K-T phase fluctuations. Therefore, the first signature is not a stringent test of the K-T transition. The second signature of the K-T transition is the nonlinear resistivity induced by finite current at $T < T_{\text{K-T}}$.[8,9,12] This nonlinear resistivity, which is a result of opposite effects from the current to the vortices and antivortices, is proportional to the quantity $(j/j_0)^{\pi K_R}$,[8,9,12] where $j_0 = T_{\text{K-T}}e/\hbar\xi_{ab}(T_{\text{K-T}})$, j is the applied current, and K_R, the stiffness of the phase fluctuations, is defined as[6,8,9] $K_R^{-1}(T)=m^*k_BT/\hbar^2 n_s^{2D}$. Theoretically,[8,9] the $\ln V$ vs $\ln I$ plot below $T_{\text{K-T}}$ shows a non-Ohmic behavior with a constant slope $N(T) \equiv 1+\pi K_R(T)$ in the small current limit. This type of power-law dependence is different from the exponential dependence $V \propto \exp(I/I_0)$, with I_0 a constant, for vortices motion due to flux depinning.[10] The third signature of the K-T transition is that since $K_R \to 2/\pi$ and n_s^{2D} is nonzero for $T \to T_{\text{K-T}}^-$, and since the superfluid density $n_s^{2D}=0$ for $T > T_{\text{K-T}}$, $N(T)$ should exhibit a universal jump at $T_{\text{K-T}}$.[12,13]

The crystal growing process and the details of the sample preparation have been described elsewhere.[14] The typical size of the single crystals was about $0.5 \times 0.4 \times 0.03$ mm^3. Contacts were made by evaporating small gold pads onto the sample, followed by subsequent bonding of gold wires to the gold pads. Eight contacts were attached to the crystal, four on the top and four on the bottom. We used the standard Montgomery arrangements and took van der Pauw corrections into account. We measured the resistance (R) versus temperature (T) for both the ab plane and c axis by applying a small ac current (typically 0.25 mA and 27.5 Hz) and using a lock-in technique. I-V characteristics were studied using dc current. A small current range (from 0 to 5 mA) was chosen in measuring the I-V characteristics in a narrow temperature interval ($92.5 < T < 94.0$ K) for both the ab plane and c axis. The temperature stability was ± 10 mK. Both the resistivity and the I-V measurements were performed on all possible combinations using the attached eight pads, so that the self-consistency of the experimental data is assured. We have also performed similar measurements on three other single crystals, and found that the I-V characteristics are all consistent with the power-law dependence, as described below.

The R vs T data show that the zero resistance for both the ab plane and the c axis occurs at the same temperature ($\simeq 92.96$ K). We now compare the ab-plane resistivity data at $T > T_{\text{K-T}}$ (see Fig. 1) to the R vs T behavior of the K-T transition. If we take the temperature where the onset of resistivity appears as $T_{\text{K-T}}$, the $\log_{10} R_{ab}$ vs $(T - T_{\text{K-T}})^{-1/2}$ plot in Fig. 1 shows a linear behavior for the temperature range $T_{\text{K-T}} < T < T_{\text{K-T}} + 0.12$ K, consistent with the K-T prediction.[6,8] The best fit shown in Fig. 1 yields $2b'(T_c^0/T_{\text{K-T}} - 1)^{1/2} = 0.14$ and $T_{\text{K-T}} = 92.97 \pm 0.01$ K. We note that other fits with $T_{\text{K-T}}$ 0.02 K away from 92.97 K fail to show a linear behavior between $\log_{10} R_{ab}$ and $1/(T/T_{\text{K-T}} - 1)^{1/2}$, and therefore the assignment of $T_{\text{K-T}} = 92.97 \pm 0.01$ K is reasonable. At higher temperatures other effects such as the Aslamazov-Larkin paraconductivity[10,11] become important, and therefore the resistivity data departs from the temperature dependence of the K-T vortices. We attribute the midpoint temperature, where the $\log_{10} R_{ab}$ data shows a clear change in the slope, to the MF superconducting temperature $T_c^0 \simeq 93.15 \pm 0.02$ K. As we shall show below, this number agrees very well with the I-V analyses.

The ab-plane I-V data at various temperatures are shown in Fig. 2. The voltage resolution is 1 μV. These data are analyzed in Fig. 3(a), where $V \propto I^{N(T)}$. We note that $N(T)$ is a well-defined constant of current for $T < T_{\text{K-T}}$, and is as large as ~ 9 at 92.66 K; in contrast, for $T > T_{\text{K-T}}$, $N(T)$ becomes current dependent: In the low-current limit, currents only contribute to the dissociation of the bound vortices, and therefore $N(T)$ jumps from 3 at $T \leq T_{\text{K-T}}$ to 1 at $T > T_{\text{K-T}}$, consistent with the third K-T signature. On the other hand, higher currents induce additional free vortices,[12,13] and the I-V behavior becomes $V \propto (I/I_0)^3 \ln[(I_0/I)]^{-1} \simeq \text{const} \times (I/I_0)^3$ at $T_{K\text{-}T} < T < T_c^0$. As the external current reaches the depairing limit, the system becomes normal and obeys the Ohmic behavior, and therefore $N(T) = 1$. Clearly our experimental data for $T \geq T_{\text{K-T}}$ [Fig. 3(a)] are consistent with the K-T

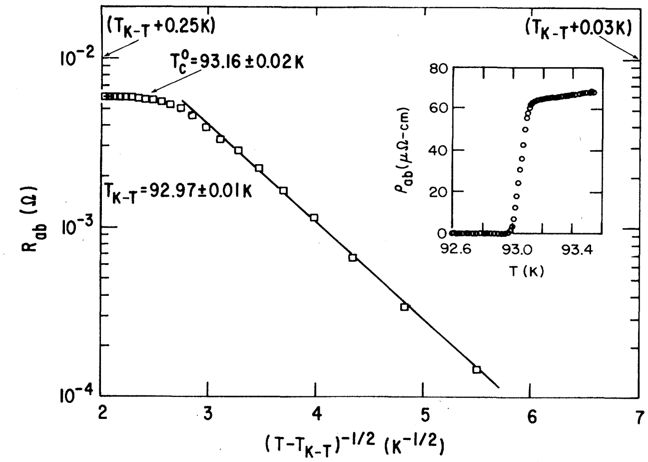

FIG. 1. The temperature dependence of the in-plane resistance is shown by the $\log_{10} R_{ab}$ vs $(T - T_{\text{K-T}})^{-1/2}$ plot, where $T_{\text{K-T}} = 92.97$ K. The linear dependence $\log_{10} R_{ab} \propto 1/(T - T_{\text{K-T}})^{1/2}$ begins to break down as temperature approaches the MF superconducting temperature ($T_c^0 = 93.15$ K ± 0.02 K). The inset shows the ab-plane resistivity transition near T_c^0.

picture. It should be emphasized that in the temperature interval $|1-t| < 2 \times 10^{-3}$, flux pinning becomes insignificant because of the large amplitude correlation length $\xi_{ab}(T)$, and the nonlinear I-V curves in such a temperature window can only be attributed to the unbinding of bound vortices. We have also analyzed our I-V data using a $\ln V$ vs I plot, and found that all the I-V curves in the plot show continuous change in the slope for all the temperatures; i.e., $V \propto \exp[I/I_0(I,T)]$, with I_0 being a complicated function of the temperature and the applied current, rather than a constant. Therefore, the exponential fit is inconsistent with any available flux-pinning theory, and our I-V data are best described by the power

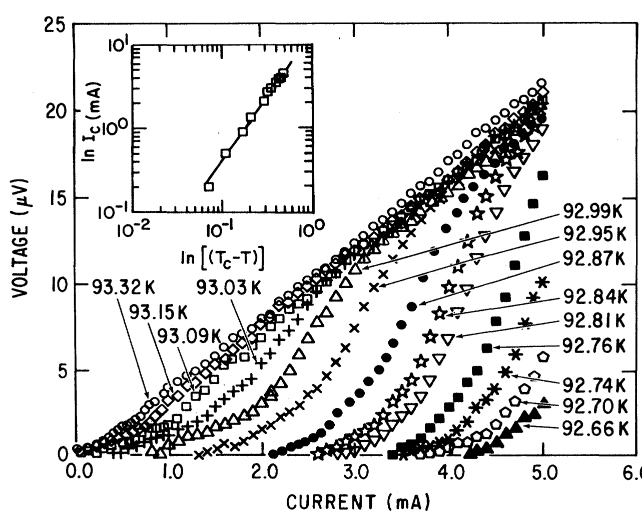

FIG. 2. I-V data for various temperatures in the ab plane. The voltage resolution is 1 μV, and the inset shows the temperature dependence of the measured critical current $I_c^{ab} \propto |1-t|^{3/2}$.

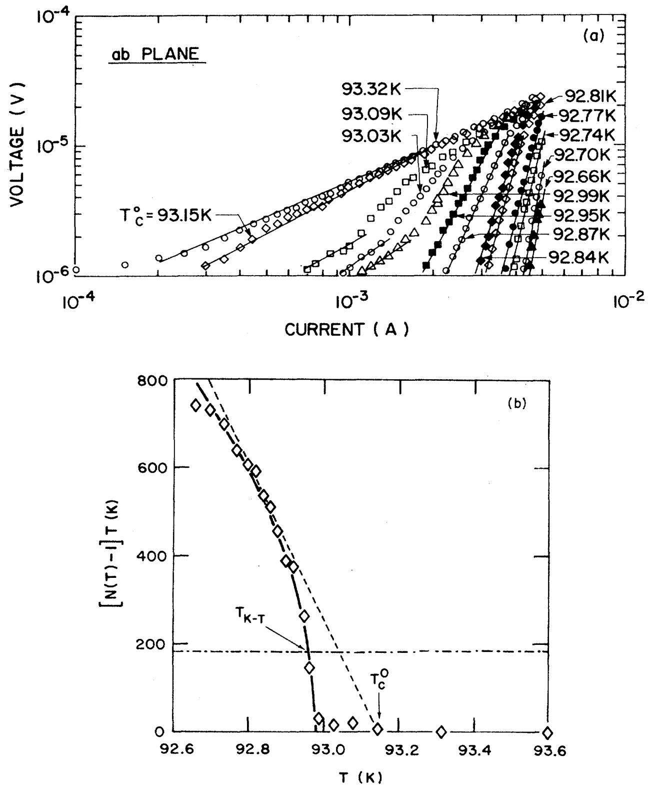

FIG. 3. (a) $\log_{10}V$ vs $\log_{10}I$ data at various temperatures in the *ab* plane. Note that the slope of each curve $[N(T)]$, where $N(T)$ is defined as $V \propto I^{N(T)}$, decreases with increasing temperature, and $N(T) \rightarrow 1$ in the high current and (or) high-temperature limit. (b) The quantity $N(T)$ obtained from (a) is analyzed by plotting $[N(T)-1]T$ vs T. The solid line is the theoretical fit $[N(T)-1]=b(T_0-T)^{1/2}$ to the data, with $b=1400$ K$^{-1/2}$ and $T_0=92.99$ K. The crossing of the solid line and the dotted line defines $T_{\text{K-T}}$, and the crossing between the dashed line and the x axis defines $T_c^0 \simeq 93.15 \pm 0.02$ K.

law $V \propto I^{N(T)}$ as the result of vortex-pair excitations. We also note that the K-T behavior in the flux flow limit does not contradict the flux creep phenomena[15–17] at temperatures sufficiently far from $T_{\text{K-T}}$ (i.e., $T \ll T_{\text{K-T}}$ or $T \gg T_{\text{K-T}}$), or for an external field $H_{c2} > H > H_{c1}$, because flux pinning and flux penetrations in these regimes become more important to the dissipation. More details regarding this point will be given elsewhere.[18]

Using the $N(T)$ values in the low-current limit, we obtain the temperature dependence of $[N(T)-1]T$ shown in Fig. 3(b). The solid line corresponds to an experimental fit to the data, which shows a temperature dependence of $[N(T)-1]T=b(T_0-T)^{1/2}$, where $b \simeq 1400$ and $T_0=92.99$ K. The crossing between the dotted line [which corresponds to $N(T)=3$] and the solid line defines K-T temperature ($T_{\text{K-T}} \simeq 92.97 \pm 0.01$ K);[9,13] and the crossing between the dashed line (which corresponds to $[N(T)-1]T=(T_c^0-T)/\epsilon(T)$) and the x axis defines the mean field $T_c^0 \simeq 93.15 \pm 0.02$ K.[13] These values are consistent with the $R(T_{\text{K-T}} < T < T_c^0)$ vs T analysis in Fig. 1. In short, the *ab*-plane transport properties are all consistent with the picture of the K-T transition, 0.2 K below the MF superconducting transition. Although indications of K-T transitions have been reported before in both ceramic and single-crystal $YBa_2Cu_3O_7$,[3] our current results provide the first clear evidence of a universal jump.

Using the characteristics of the Kosterlitz-Thouless transition, we can estimate the "effective thickness" (l_c) of the sample by the following consideration. For a thin-film superconductor with a large κ ($\equiv \lambda/\xi$, where λ is the penetration depth and ξ the correlation length), Beasley, Mooij, and Orlando[7] have shown that

$$\frac{T_c^0}{T_{\text{K-T}}}=1+0.173\epsilon\left[\frac{\rho_{ab}/l_c}{\hbar/e^2}\right], \quad (T_c^0-T_{\text{K-T}})/T_{\text{K-T}} \ll 1 \,. \quad (1)$$

Fiory *et al.*[4] have estimated $\epsilon \simeq 4.6$ for $YBa_2Cu_3O_7$ thin film at $T \simeq T_{\text{K-T}}$. With the *ab*-plane resistivity $\rho_{ab}(T=T_c^0)=65$ $\mu\Omega$ cm, $\epsilon \simeq 4.6$, $T_c^0=93.15$ K, and $T_{\text{K-T}}=92.97$ K, we obtain $l_c/\epsilon \simeq 140$ Å at $T_{\text{K-T}}$. In a very thin film where the c-axis correlation length is larger than the thickness of the film, l_c in Eq. (1) is identical to the film thickness. On the other hand, in thick films or bulk single crystals, the effective thickness l_c should scale with the c-axis coherence length $\xi_c(T)$.[18] Since $\xi_c(t) \propto (1-t)^{-1/2}$, and $\epsilon \rightarrow 1$ for $T \rightarrow 0$,[6] we obtain $l_c(0) \simeq 6$ Å. It is interesting to note that the $l_c(0)$ value is about the spacing between two CuO_2 planes (5.85 Å) in $YBa_2Cu_3O_7$, and that the small separation between T_c^0 and $T_{\text{K-T}}$ is consistent with the relatively small sheet resistance ($R_\square \simeq 50$ Ω) in the $YBa_2Cu_3O_7$ system.

Given a correlation length l_c, the supercarrier effective mass m^* may be estimated from the unbinding energy of the paired vortices,[6,8] i.e.,

$$k_B T_{\text{K-T}}=\frac{\pi\hbar^2}{4}\left[\frac{n_s^{2D}}{m^*_{\text{K-T}}}\right], \quad T \rightarrow T_{\text{K-T}}^- \,. \quad (2)$$

If we substitute n_s^{2D} by the supercarrier density renormalized over the thickness l_c; that is, $n_s^{2D}(T)=n_s^{3D}(T)l_c(T)$, the effective mass yields the following formula:

$$m^*_{\text{K-T}}/m_e=7.5\times10^{-14}(\text{cm}^{-2})n_s^{2D}(T_{\text{K-T}}) \,. \quad (3)$$

Assuming that $n_s^{2D}(t) \propto (1-t)^{1/2}$ for $t=T/T_c^0 \lesssim 1$,[10] and $n_s^{3D}(0) \simeq 10^{22}$ cm^{-3},[4] we obtain $n_s^{2D}(0)=6\times10^{14}$ cm^{-2} and $m^*_{\text{K-T}} \simeq 9m_e$. It should be noted that the absolute value of the effective mass depends on the accuracy of the measured n_s^{3D}. The physical meaning of $m^*_{\text{K-T}}$ may be the mass unit of the supercarriers renormalized within a thickness l_c in a quasi-two-dimensional system. The large two-dimensional effective mass ($m^*_{\text{K-T}} \simeq 9m_e$) is suggestive of a strong in-plane many-body interaction.[19]

We now compare the consistency of our values for $m^*_{\text{K-T}}$ and n_s^{2D}, which are based on Kosterlitz-Thouless theory, to those obtained from other measurements, such as the London penetration depth[4,20] λ_L and the Sommerfeld constant[21] γ. Using the measured in-plane penetration depth ($\lambda_{ab} \simeq 1500$ Å),[4,20] and the formula $\lambda_L^2=c^2m^*_{3D}/4\pi n_s^{3D}e^2$, and assuming $n_s^{3D}=10^{22}$ cm^{-3}, we obtained $m^*_{ab} \simeq 8m_e$, in good agreement with our estimate ($m^*_{\text{K-T}} \simeq 9m_e$). We note that the effective penetration depth $\Lambda \equiv 2\lambda_{ab}^2/l_c \simeq 0.1$ mm at $T=0$, which is about the size of our sample. Therefore, the K-T behavior is expected in the length scale $r \ll \Lambda$.[7] We also considered the Sommerfeld constant γ for a stack of two-dimensional systems with a spacing d_0 between them; that is, $\gamma=(\pi k_B^2/3\hbar^2)(m_{2D}^*/d_0)$, and employed the experimental value $\gamma\simeq 10$ mJ/(mole Cu K^{-2}).[21] The effective mass thus estimated is $m^*/m_e\simeq 10$ if we take $d_0=l_c(0)\simeq 6$ Å. This number is again consistent with $m_{\text{K-T}}^*$.

Next, we discuss the superconducting properties along the c axis. As shown in Fig. 4, $N(T)$ vs T shows that $1<N(T)\sim 3$ for temperatures below the mean-field superconducting temperature T_c^0. The non-Ohmic I-V curves suggest current-induced flux flow along the c axis.[18,22] On the other hand, for $T>T_c^0$, the c-axis I-V curves restore the Ohmic behavior $[N(T)=1]$. The anomalous jump of $N(T)=3$ at 92.98 K may be attributed to current redistribution effects[14] from the ab plane to the c axis, due to the onset of the in-plane resistivity at $T_{\text{K-T}}$. The $N(T)$ vs T plot for the c-axis data is significantly different from that of the ab plane. Assuming that the resistivity at $T<T_c^0$ in the zero magnetic field limit is basically thermally activated, we anticipate the onset of resistance along the c axis to follow a temperature dependence $R_c(T)\propto\exp[-U(T)/k_BT]$, where $U(T)$ is a pinning potential.[22] The best fit to the experimental data shows that $U(T)\propto(T_c^0-T)^2$. More details about the pinning mechanism along the c axis will be discussed elsewhere.[18] In short, the c-axis superconductivity shows distinct contrasts to those in the ab plane, and the electrical properties along the c axis may be treated as those of superconducting weak links, consistent with the observed K-T transition in the ab plane.

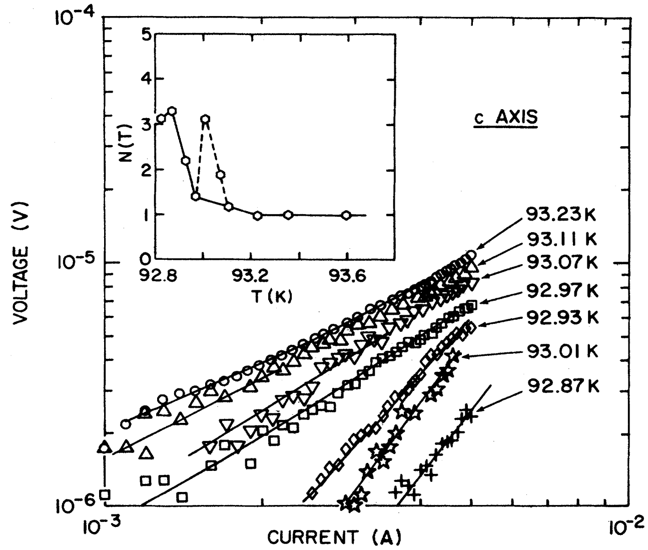

FIG. 4. $\log_{10}V$ vs $\log_{10}I$ data of the c axis at various temperatures. The inset shows a decrease of the power $N(T)$ with the increasing temperature, and $N(T)=1$ for $T>T_c^0$. At $T\simeq T_{\text{K-T}}$, an anomalous jump of $N(T)=3$ is probably due to the current redistribution effect from the ab plane.

In conclusion, we have studied the electrical transport properties of single-crystal $YBa_2Cu_3O_7$ near the superconducting transition temperature. Experimental results indicate Kosterlitz-Thouless–type behavior in the ab plane, and therefore manifest the quasi-two-dimensional nature of the superconducting oxide. We have estimated the c-axis correlation length $l_c(T=0)\simeq 6$ Å and the in-plane effective mass of the quasi-two-dimensional supercarriers $(\sim 10m_e)$ based on the Kosterlitz-Thouless theory. The large effective mass implies a strong many-body interaction among the supercarriers confined in a quasi-two-dimensional sheet with an effective thickness l_c. In contrast, the temperature dependence of the resistivity and the I-V characteristics along the c axis are consistent with a thermally actived flux-flow model. This work has provided experimental evidences for low dimensionality and strong many-body interaction in superconducting $YBa_2Cu_3O_7$, and these are believed to be essential to the pairing mechanism.

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