Low-temperature nodal-quasiparticle transport in lightly doped YBa$_2$Cu$_3$O$_y$ near the edge of the superconducting doping regime

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In-plane transport properties of nonsuperconducting YBa$_2$Cu$_3$O$_y$ ($y = 6.35$) are measured using high-quality untwinned single crystals. We find that both the $a$- and $b$-axis resistivities show log$(1/T)$ divergence down to 80 mK, and accordingly the thermal conductivity data indicate that the nodal quasiparticles are progressively localized with lowering temperature. Hence, both the charge and heat transport data do not support the existence of a “thermal metal” in nonsuperconducting YBa$_2$Cu$_3$O$_y$, as opposed to a recent report by Sutherland et al. [Phys. Rev. Lett. 94, 147004 (2005)]. Besides, the present data demonstrate that the peculiar log$(1/T)$ resistivity divergence of cuprate is not a property associated with high-magnetic fields.

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The electronic properties of underdoped cuprates have been extensively studied in recent years, with the aim of understanding how the strong electron correlations in these materials manifest themselves and lead to high-$T_c$ superconductivity. Most notably, it has been elucidated that the electronic density of states near the Fermi energy $E_F$ is gradually suppressed from above $T_c$ in underdoped cuprates, causing a “pseudogap” in the normal state. The pseudogap opens anisotropically in the Brillouin zone and has four nodes along the zone diagonals, similar to the $d$-wave superconducting (SC) gap. Furthermore, the pseudogap is formed through partial destruction of the Fermi surface from the antinodes, leaving “Fermi arcs” near the zone diagonals. These Fermi arcs are observed not only in the normal state of moderately underdoped superconductors but also in lightly doped nonsuperconducting (NSC) samples, which has lead to the idea that the Mott-insulating state of parent cuprates turns into a metal-like state through creation of nodal quasiparticles on the Fermi arcs. Intriguingly, the “metallic” transport supported by the Fermi arcs in lightly doped cuprates has surprisingly good mobility at moderate temperatures and shows a behavior reminiscent of ordinary Fermi liquids ($T^2$ inelastic scattering rate and $T$-independent Hall coefficient).

It should be noted, however, that those nodal quasiparticles in lightly doped cuprates are bound to be localized in the $T → 0$ limit, giving rise to variable-range hopping (VRH) behavior at low temperatures. The nodal quasiparticles in the SC state, on the other hand, are known to be delocalized in the $T → 0$ limit and can carry heat, which has been documented by the measurements of the thermal conductivity $κ$ at milli-Kelvin temperatures that find a finite $T$-linear term expected for fermionic excitations throughout the superconducting doping regime of various cuprates. Notably, it is also known that the normal state of sufficiently underdoped cuprates becomes “insulating” in the $T → 0$ limit when the superconductivity is suppressed with a high magnetic field. In fact, the magnetic-field ($H$) dependence of $κ$ in the SC state is indicative of a magnetic-field-induced quasiparticle localization at sufficiently low doping, which seems to be consistent with the insulating normal state under high magnetic fields.

However, Sutherland et al. have recently argued that the ground state of lightly-doped YBa$_2$Cu$_3$O$_y$ (YBCO) in the NSC regime is a “thermal metal”, proposing that the nodal quasiparticles can remain delocalized in the underdoped NSC regime of a clean cuprate. This claim implies that $ρ_c$ falls into the NSC doping regime in YBCO, which questions the universality of the hidden MI transition in the SC regime. Naturally, if the nodal quasiparticles indeed remain delocalized in a NSC sample, one should be able to confirm the metallic nature by measuring the resistivity, since there is no superconductivity that short-circuits the dc charge transport; unfortunately, Sutherland et al. relied only on heat transport measurements for drawing their conclusion. Indeed, the intrinsic resistivity behavior of YBCO near the edge of the SC doping regime has been notoriously difficult to be elucidated, mostly because the resistivity data are easily contaminated by filamentary superconductivity at such doping.

In this paper, we critically examine whether there is really a thermal metal phase in clean YBCO in the NSC regime, by measuring both the charge and heat transport properties on untwinned single crystals down to 70–80 mK. We have succeeded in preparing samples that are free from filamentary superconductivity and found that at $y = 6.35$, which is very close to the SC doping regime ($y ≥ 6.40$), the temperature dependences of the $a$- and $b$-axis resistivities ($ρ_a$ and $ρ_b$) are both well described by log$(1/T)$ in zero field, which obviously indicates that the
nodal quasiparticles are localized in the zero-temperature limit. Also, this charge-transport behavior signifies that the nodal quasiparticles are *inelastically* scattered down to the lowest temperature, and hence dictates that one should never assume the electronic thermal conductivity to be linear in $T$, an assumption that was employed in the analysis by Sutherland et al.\textsuperscript{12} but is only valid in the elastic scattering regime of fermionic excitations. In fact, the behavior of the $a$- and $b$-axis thermal conductivities ($\kappa_a$ and $\kappa_b$) indicates that the nodal quasiparticles are progressively localized upon lowering temperature.

The high-quality single crystals of YBa$_2$Cu$_3$O$_y$ are grown by a flux method using Y$_2$O$_3$ crucibles to avoid inclusion of impurity atoms\textsuperscript{20,21} using elaborate procedures to reliably control the oxygen content $y$ described in Ref. 10; the samples are carefully annealed and detwinned. Note that our samples are quenched after the high-temperature annealing to avoid long-range oxygen ordering, although formations of short-range Cu-O chain fragments always take place in low-doped YBCO samples at room temperature after quenching — this is the cause of the “room-temperature annealing” (RTA) effect\textsuperscript{12,13}. The $y$ values of the samples reported here are 6.45, 6.40, and 6.35, for which the zero-resistivity $T_c$’s are 20, 7.5, and 0 K, respectively. The SC samples ($y = 6.45$ and 6.40) are measured after one week of RTA, after which the change of $T_c$ almost saturates. On the other hand, the $y = 6.35$ samples are measured within one day of RTA, because the one-week RTA tends to cause a filamentary superconductivity which contaminates the resistivity data below 2 K; thus, the present $y = 6.35$ samples can be considered to be essentially oxygen-order free. The resistivity and thermal conductivity are independently measured by using the conventional four-probe method and a steady-state technique\textsuperscript{20,21}, respectively.

The inset to Fig. 1(a) shows the temperature dependences of $\rho_a$ and $\rho_b$ for $y=6.35$ below 300 K, where one can see that the resistivity shows a metallic behavior down to 70 K (40 K) along the $a$ ($b$) axis, but a steep upturn sets in at lower temperature. The main panel of Fig. 1(a) zooms in on the data below 20 K; here, one can clearly see that both $\rho_a$ and $\rho_b$ are apparently diverging at low temperature and it is difficult to assert some definite resistance value for their $T \to 0$ limit. In fact, as shown in Fig. 1(b), the $T$ dependences of both $\rho_a$ and $\rho_b$ are well described by $\log(1/T)$ below ~20 K down to 80 mK (more than two decades), and the $\log(1/T)$ dependence mathematically suggests that the resistivity grows infinitely large for $T \to 0$. Note that this behavior is the same as that found in the normal state of underdoped cuprate superconductors under high magnetic fields\textsuperscript{13,16}, but this is the first time that a clear $\log(1/T)$ behavior is observed in zero magnetic field: When $T_c$ is suppressed to zero by Zn doping in underdoped samples, the resistivity shows a divergence quicker than $\log(1/T)$\textsuperscript{22,23}. Incidentally, we found that application of a 16-T magnetic field causes negligible change in the resistivity data of the present samples. Hence, the present data demonstrate that the $\log(1/T)$ resistivity divergence of cuprates is *not* a property associated with high-magnetic fields; also, the $\log(1/T)$ behavior in clean YBCO suggests that this peculiar behavior is not merely a result of disorder-induced localization but is fundamentally related to the strong-correlation physics.

Figure 1(c) shows the $T$ dependence of the in-plane anisotropy, $\rho_a/\rho_b$, for $y=6.35$. As we have reported previously\textsuperscript{22}, $\rho_a/\rho_b$ grows to as large as ~2.5 at this doping, even though the orthorhombicity is very weak (0.2%) and is about to disappear — this is arguably the most convincing evidence that the electrons self-organize into a macroscopically anisotropic state (such as a nematic charge-stripe phase\textsuperscript{25}) in lightly-doped YBCO\textsuperscript{16}. In Fig. 1(c), one can see that $\rho_a/\rho_b$ becomes nearly $T$-independent when the resistivity is showing the $\log(1/T)$ divergence. This is rather reminiscent of the behavior of the out-of-plane anisotropy, $\rho_c/\rho_{ab}$, in underdoped LSCO, which also shows a saturation when the resistivities individually show the $\log(1/T)$ divergence\textsuperscript{12}.

Figure 2 shows the corresponding $\kappa_a$ and $\kappa_b$ data for $y=6.35$. In Fig. 2(a), $\kappa/T$ is plotted versus $T^2$, which would allow one to separate the electronic contribution $\kappa_e$ ($\sim T$ in the elastic-scattering regime) from the phononic contribution $\kappa_p$ ($\sim T^3$ in the boundary-scattering regime), provided that the data at the lowest temperatures [usually below 100–200 mK (Refs. \textsuperscript{3,15,26})] fall onto a straight line; however, one can easily see in Fig. 2(a) that both the $\kappa_a$ and $\kappa_b$ data are continuously curved in this plot down to our lowest temperature, 70 mK, and one cannot make a linear fitting for any reasonably extended temperature range. In fact, the data look as if $\kappa/T$ is heading to zero at $T=0$. Note
that, by choosing some fractional power of $T$ for the horizontal axis, one could make a plot that shows an almost linear behavior [Fig. 2(b)], and this fact corroborates the observation that the data are nowhere linear in the ordinary $\kappa/T$ vs $T^2$ plot. Actually, as we discuss later, when the resistivity shows a log(1/$T$) divergence and hence an inelastic scattering process is affecting the electron transport, there is no reason to believe that the electronic part of $\kappa/T$ is constant, and therefore it is not meaningful to play around with the data to try to separate $\kappa_p$ by making a plot like Fig. 2(b). In other words, the absence of the linear behavior in the $\kappa/T$ vs $T^2$ plot in the present case does not mean that the boundary-scattering regime of phonons (where $\kappa_p \sim T^3$) is never achieved nor the proper power for $\kappa_p$ is different from 3, but it means that $\kappa_e$ never behaves as $\sim T$ due to the log(1/$T$) localization behavior. We note that in our experiments both the resistivity and thermal conductivity results for $y=6.35$ are confirmed to be essentially reproduced in at least one more set of samples.

It should be remarked that there is a notable difference in the thermal conductivity behavior between SC samples and NSC samples; namely, for SC samples there is always a finite range of temperature where the data are linear in the $\kappa/T$ vs $T^2$ plot with a finite intercept at $T=0$, as shown in Fig. 3(a), while for NSC samples the data in such a plot are curved all the way down to the lowest $T$. To make it easier to judge the rationality of the linear fittings for the SC samples, Fig. 3(b) shows the difference between the data and the fits; here, one can see that the linear fitting is very reasonable below 0.013 and 0.010 K$^2$ (i.e., 115 and 100 mK) for $y = 6.45$ and 6.40, respectively. Physically, the observed change in the thermal conductivity behavior across the superconductivity boundary is a manifestation of the fact that the nodal quasiparticles are localized on the NSC side of this boundary, while they are delocalized in the SC state.

In passing, we note that the linear fittings in Fig. 3 for $y = 6.45$ and 6.40 are very consistent with what is expected for $\kappa_p$: In the boundary-scattering regime, $\kappa_p$ is given by $\frac{1}{3}(v_p l_p)^2 T^3$, with $\beta$ the phonon specific heat coefficient, $\langle v_p \rangle$ the averaged sound velocity, and $l_p$ the phonon mean-free path; in perfect crystals $l_p$ takes the maximum value $1.12\bar{w}$ with $\bar{w}$ the geometric mean width of the sample. Using the parameters for YBCO cited in Ref. 8 and $\bar{w}$ of 0.356 (0.245) mm for $y = 6.45$ (6.40), we obtain $l_p/1.12\bar{w}$ of 0.85 (0.92).

Let us now discuss the most important question of whether the ground state at $y=6.35$ is a metal or an insulator. For this discussion, it is crucial to examine the meaning of the log(1/$T$) resistivity divergence. Although this behavior is not completely equivalent to the log $T$ behavior of the conductivity $\sigma$ in 2D weak localization,[27] it is instructive to recall the basic physics behind the weak localization. According to the scaling theory of weak localization,[26] the inelastic diffusion length $L_i$ (which grows upon lowering $T$) sets the length scale to determine the size-dependent conductivity $g(L_i)$, which depends logarithmically on $L_i$ and vanishes for $L_i \rightarrow \infty$; this dependence on $L_i$ is the essential source of the log $T$ behavior.[26] Therefore, when the log $T$ weak localization behavior is observed, the transport is governed by the inelastic scattering process and one can never assume $\kappa_e$ to behave as $\sim T$ as in the elastic-scattering regime. In fact, since all the electrons are eventually localized in the log $T$ regime, $\kappa_e/T$ is bound to vanish for $T \rightarrow 0$.

In the case of the log(1/$T$) resistivity behavior in
cuperates, although the exact mechanism to produce the logarithmic dependence would be different from the weak localization, the system is certainly not in the elastic-scattering regime and some inelastic process continues to be effective for \( T \to 0 \), causing the ground state at \( T=0 \) to be a correlated insulator. This means that it makes best sense to interpret our data for \( y=6.35 \) to be indicative of a progressive localization of electrons and vanishing \( \kappa_e/T \) for \( T \to 0 \). Also, the \( \log(1/T) \) resistivity behavior implies that it is not reasonable to assert \( \kappa_e/T \) to be constant and hence the \( \kappa/T \)-vs-\( T \) analysis like that in Fig. 2(b) is essentially meaningless. In this context, it is also pointless to examine the validity of the Wiedemann-Franz (WF) law in the \( \log(1/T) \)-insulating regime, because the WF law holds only in the elastic-scattering regime.\(^{20}\)

Having discussed that the data for \( y=6.35 \) are indicative of a vanishing \( \kappa_e/T \) for \( T \to 0 \), we can now draw a diagram for the doping dependence of the \( T=0 \) residual electronic term, \( \kappa_0/T \), as shown in Fig. 3(c), where the data obtained in our previous work\(^{10}\) for \( 6.50 \leq y \leq 7.00 \) are also plotted. Altogether, there is a good systematics in the doping dependence of \( \kappa_0/T \), which is essentially consistent with that for LSCO\(^{12}\) giving confidence that the simultaneous disappearance of the superconductivity and delocalized nodal quasiparticles is a fundamental nature of the cuprates. The present result also indicates that even in the cleanest cuprate YBCO the ground state of the lightly-doped NSC regime is an insulator, though the nature of the insulating state, which is characterized by the \( \log(1/T) \) resistivity divergence here, is different from that in lightly doped LSCO where it is due to the disorder-driven localization.\(^{5}\)

Lastly, let us briefly discuss the possibility that, while the charges are localized and cannot give rise to a thermal metal phase in the NSC regime of YBCO, some exotic fermionic excitations might be alternatively responsible for a thermal metal in the charge insulating state. One can easily see that this scenario is very unlikely, because, as shown in Fig. 3(a), \( \kappa_0/T \) decreases notably and systematically from \( y=6.45 \) to \( 6.35 \), which speaks against the idea that some new fermions start to carry heat for \( y \leq 6.35 \) and contribute a sizable \( \kappa_0/T \).

In conclusion, we find that in clean, unwinned single crystals of nonsuperconducting (NSC) YBCO at \( y = 6.35 \), the temperature dependences of both \( \rho_a \) and \( \rho_b \) show \( \log(1/T) \) divergence in zero magnetic field, which indicates that the nodal quasiparticles in the NSC regime are localized for \( T \to 0 \) and that they are inelastically scattered down to the lowest temperature. The latter means that the electronic thermal conductivity \( \kappa_e \) cannot be linear in \( T \) even at the lowest temperature, and accordingly the data for \( \kappa_a \) and \( \kappa_b \) at \( y = 6.35 \) are indicative of a \( T \)-dependent \( \kappa_e/T \) that appears to vanish for \( T \to 0 \). Hence, the critical examination of the heat transport at \( y = 6.35 \) in the light of the charge-transport behavior leads to a conclusion that the ground state of clean underdoped cuprate in the NSC regime is not a thermal metal but is a peculiar insulator characterized by the \( \log(1/T) \) resistivity divergence.

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