PHYSICAL REVIEW B **72**, 100502(R) (2005)

Low-temperature nodal-quasiparticle transport in lightly doped $YBa_2Cu_3O_y$ near the edge of the superconducting doping regime

X. F. Sun,* Kouji Segawa, and Yoichi Ando†
Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan

(Received 24 June 2005; published 14 September 2005)

In-plane transport properties of nonsuperconducting $YBa_2Cu_3O_y$ (y=6.35) are measured using high-quality untwinned single crystals. We find that both the a- and b-axis resistivities show $\ln(1/T)$ 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 $YBa_2Cu_3O_y$, as opposed to a recent report by Sutherland *et al.* [Phys. Rev. Lett. **94**, 147004 (2005)]. Besides, the present data demonstrate that the peculiar $\ln(1/T)$ resistivity divergence of cuprate is *not* a property associated with high magnetic fields.

DOI: 10.1103/PhysRevB.72.100502 PACS number(s): 74.25.Fy, 74.25.Dw, 74.72.Bk

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.[1] 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.[1,2] The pseudogap opens anisotropically in the Brillouin zone and has four nodes along the zone diagonals, similarly 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.[3] These Fermi arcs are observed not only in the normal state of moderately underdoped superconductors but also in lightly doped nonsuperconducting (NSC) samples,[4] which has led 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.[4,5] Intriguingly, the "metallic" transport supported by the Fermi arcs in lightly doped cuprates has surprisingly good mobility at moderate temperatures[6] and shows a behavior reminiscent of ordinary Fermi liquids (T^2 inelastic scattering rate and T-independent Hall coefficient).[5]

It should be noted, however, that those nodal quasiparticles in lightly doped cuprates are bound to be localized in the $T\rightarrow 0$ limit, giving rise to variable-range hopping (VRH) behavior at low temperature.[6] The nodal quasiparticles in the SC state, on the other hand, are known to be *delocalized* in the $T\rightarrow 0$ limit and can carry heat,[7,8] 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.[9–12] Notably, it is also known that the normal state of sufficiently underdoped cuprates becomes "insulating" in the $T\rightarrow 0$ limit when the superconductivity is suppressed with a high magnetic field;[13–16] 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,[10,11,17] which seems to be consistent with the insulating normal state under high magnetic fields. Hence, at low doping, it appears that the nodal quasiparticles are delocalized only in the SC state, and either suppression of the superconductivity by high magnetic fields or underdoping into the NSC regime causes the nodal quasiparticles to be localized. This picture is consistent with the idea that there is a "hidden" metal-insulator (MI) transition at a critical doping p_c in the SC doping regime, and below p_c the nodal quasiparticles are localized in the absence of superconductivity.

However, Sutherland *et al.*[12] have recently argued that the ground state of lightly doped $YBa_2Cu_3O_y$ (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 p_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.[12] Indeed, the intrinsic resistivity behavior of YBCO near the edge of the SC doping regime has been notoriously difficult to elucidate, 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 the NSC regime of clean YBCO 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 ($y\geqslant 6.40$), the temperature dependences of the a- and b-axis resistivities (ρ_a and ρ_b) are both well described by $\ln(1/T)$ 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.*[12] but is only valid in the elastic scattering regime of fermionic excitations. In fact, the behav-

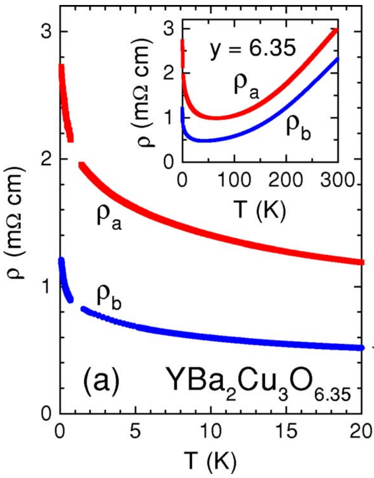

FIG. 1. (Color online) (a) T dependences of zero-field ρ_a and ρ_b for $y=6.35$ below 20 K; inset shows the data up to 300 K. (b) Semilog plot of the data to show the $\ln(1/T)$ divergence. (c) T dependence of the in-plane anisotropy, ρ_a/ρ_b. The slight discontinuity in the data across 1 K is due to the use of a very small excitation current in the dilution-fridge measurements.

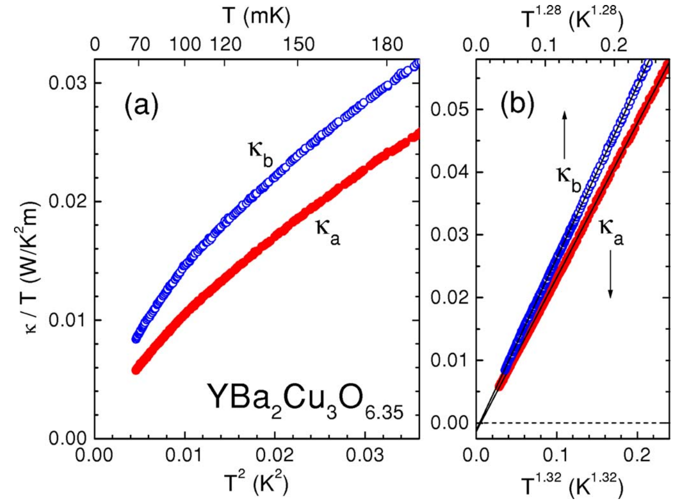

FIG. 2. (Color online) (a) T dependences of κ_a and κ_b for $y=6.35$ down to 70 mK, plotted in κ/T vs T^2. (b) κ/T vs T^α analysis of κ_a and κ_b for $y=6.35$ to artificially bring out a linear behavior.

ior of the a- and b-axis thermal conductivities (κ_a and κ_b) indicates that the nodal quasiparticles are progressively localized upon lowering temperature.

The high-quality single crystals of $YBa_2Cu_3O_y$ are grown by a flux method using Y_2O_3 crucibles to avoid inclusion of impurity atoms;[18,19] we have documented[10] that our crystals are of comparable cleanliness to the best crystals grown in $BaZrO_3$ crucibles.[20,21] Using elaborate procedures to reliably control the oxygen content y described in Ref. 19, 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.[12,18] 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 that 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[6] and a steady-state technique,[9,10] respectively.

The inset to Fig. 1(a) shows the temperature dependences of ρ_a and ρ_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 ρ_a and ρ_b are apparently diverging at low temperature and it is difficult to assert some definite resistivity value for their $T\rightarrow 0$ limit. In fact, as shown in Fig. 1(b), the T dependences of both ρ_a and ρ_b are well described by $\ln(1/T)$ below $\sim$20 K down to 80 mK (more than two decades), and the $\ln(1/T)$ dependence mathematically suggests that the resistivity grows infinitely large for $T\rightarrow 0$. Note that this behavior is the same as that found in the normal state of underdoped cuprate superconductors under high magnetic fields,[13,16] but a clear $\ln(1/T)$ behavior was never observed before in zero magnetic field: When T_c is suppressed to zero by Zn doping in underdoped samples, the resistivity shows a divergence quicker than $\ln(1/T)$.[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 $\ln(1/T)$ resistivity divergence of cuprates is *not* a property associated with high magnetic fields; the $\ln(1/T)$ behavior in clean YBCO also 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 resistivity anisotropy, ρ_a/ρ_b, for $y=6.35$. As we have reported previously,[24] ρ_a/ρ_b grows to be as large as $\sim$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[25]) in lightly doped YBCO.[24] In Fig. 1(c), one can see that ρ_a/ρ_b becomes nearly T independent when the resistivity is showing the $\ln(1/T)$ divergence. This is rather reminiscent of the behavior of the out-of-plane anisotropy, ρ_c/ρ_{ab}, in underdoped $La_{2-x}Sr_xCuO_4$ (LSCO), which also shows a saturation when the resistivities individually show the $\ln(1/T)$ divergence.[13]

Figure 2 shows the corresponding κ_a and κ_b data for $y=6.35$. In Fig. 2(a), κ/T is plotted versus T^2, which would allow one to separate the electronic contribution κ_e ($\sim T$ in the elastic-scattering regime) from the phononic contribution κ_p ($\sim T^3$ in the boundary-scattering regime), provided that the data at the lowest temperatures [usually below 100–200 mK (Refs. 8–11 and 26)] fall onto a straight line; however, one can easily see in Fig. 2(a) that both the κ_a and κ_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 κ/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 κ/T vs T^2 plot. Actually, as we discuss later, when the resistivity shows a $\ln(1/T)$ divergence and hence an inelastic scattering process is affecting the electron transport, there is no reason to believe that the electronic part of κ/T is constant, and therefore it is not meaningful to play around with the data to try to separate κ_p by making a plot like Fig. 2(b). In other words, the absence of the linear behavior in the κ/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 that the proper power for κ_p is different from 3,[12] but it does mean that κ_e never behaves as $\sim T$ due to the $\ln(1/T)$ 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 κ/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(a) for $y=6.45$ and 6.40 are very consistent with what is expected for κ_p. In the boundary-scattering regime, κ_p is given by $\frac{1}{3}\beta\langle v_{ph}\rangle l_{ph}T^3$, with β the phonon specific heat coefficient, $\langle v_{ph}\rangle$ the averaged sound velocity, and l_{ph} the phonon mean free path; in perfect crystals l_{ph} 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 our $y=6.45$ (6.40) sample, we obtain $l_{ph}/1.12\bar{w}$ 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 $\ln(1/T)$ resistivity divergence. Although this behavior is not completely equivalent to the $\ln T$ behavior of the conductivity σ in two-dimensional weak localization,[27] it is instructive to recall the basic physics behind the weak localization.

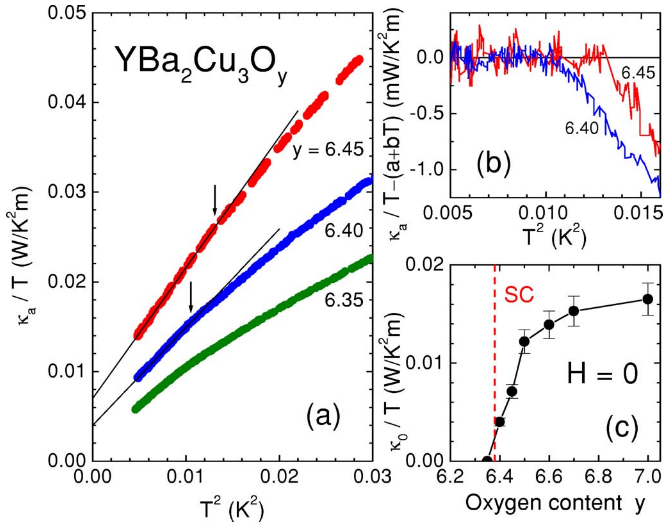

FIG. 3. (Color online) (a) κ_a/T vs T^2 plot for $y=6.45$, 6.40, and 6.35; the solid lines are the straight-line fits to the lowest-temperature data of the SC samples, and arrows mark the temperature below which the fitting is good. (b) The difference between the κ_a/T data and the linear fittings shown in (a) for the two dopings. (c) The doping dependence of κ_0/T at $H=0$ determined in the present paper and in Ref. 10; note that all the data are for the *a* axis (perpendicular to the chains).

According to the scaling theory of weak localization,[27] 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 $L_i \to \infty$; this dependence on L_i is the essential source of the $\ln T$ behavior.[28] Therefore, when the $\ln T$ weak localization behavior is observed, the transport is governed by the inelastic scattering process and one can never assume κ_e to behave as $\sim T$ as in the elastic-scattering regime. In fact, since all the electrons are eventually localized in the $\ln T$ regime, κ_e/T is bound to vanish for $T\to 0$.

In the case of the $\ln(1/T)$ resistivity behavior in cuprates, 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 the best sense to interpret our data for $y=6.35$ to be indicative of a progressive localization of electrons and vanishing κ_e/T for $T\to 0$. Also, the $\ln(1/T)$ resistivity behavior implies that it is not reasonable to assert κ_e/T to be constant and hence the κ/T vs T^α 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 $\ln(1/T)$-insulating cuprates, because the WF law holds only in the elastic-scattering regime.[29]

Having discussed that the data for $y=6.35$ are indicative of a vanishing κ_e/T for $T\to 0$, we can now draw a diagram for the doping dependence of the $T=0$ residual electronic term, κ_0/T, as shown in Fig. 3(c), where the data obtained in our previous work[10] for $6.50 \leqslant y \leqslant 7.00$ are also plotted. Altogether, there is a good systematics in the doping dependence of κ_0/T, which is essentially consistent with that for LSCO,[9] 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 $\ln(1/T)$ resistivity divergence here, is different from that in lightly doped LSCO, where it is due to the disorder-driven localization.[6]

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), κ_a/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 κ_0/T.

In conclusion, we find that in clean, untwinned single crystals of nonsuperconducting (NSC) YBCO at $y=6.35$, the temperature dependences of both ρ_a and ρ_b show $\ln(1/T)$ 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 κ_e cannot be linear in T even at the lowest temperature, and accordingly the data for κ_a and κ_b at $y=6.35$ are indicative of a T-dependent κ_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 $\ln(1/T)$ resistivity divergence.

We thank Louis Taillefer for useful discussions and sharing preprints prior to publication. This work was supported by the Grant-in-Aid for Science provided by the Japanese Society for the Promotion of Science.

---

*Electronic address: ko-xfsun@criepi.denken.or.jp
†Electronic address: ando@criepi.denken.or.jp

[1] J. Orenstein and A. J. Millis, Science **288**, 468 (2000).
[2] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[3] M. R. Norman, H. Ding, M. Randeria, J. C. Campuzano, T. Yokoya, T. Takeuchi, T. Takahashi, T. Mochiku, K. Kadowaki, P. Guptasarma, and D. G. Hinks, Nature (London) **392**, 157 (1998).
[4] T. Yoshida, X. J. Zhou, T. Sasagawa, W. L. Yang, P. V. Bogdanov, A. Lanzara, Z. Hussain, T. Mizokawa, A. Fujimori, H. Eisaki, Z.-X. Shen, T. Kakeshita, and S. Uchida, Phys. Rev. Lett. **91**, 027001 (2003).
[5] Y. Ando, Y. Kurita, S. Komiya, S. Ono, and K. Segawa, Phys. Rev. Lett. **92**, 197001 (2004).
[6] Y. Ando, A. N. Lavrov, S. Komiya, K. Segawa, and X. F. Sun, Phys. Rev. Lett. **87**, 017001 (2001).
[7] A. C. Durst and P. A. Lee, Phys. Rev. B **62**, 1270 (2000).
[8] L. Taillefer, B. Lussier, R. Gagnon, K. Behnia, and H. Aubin, Phys. Rev. Lett. **79**, 483 (1997).
[9] J. Takeya, Y. Ando, S. Komiya, and X. F. Sun, Phys. Rev. Lett. **88**, 077001 (2002).
[10] X. F. Sun, K. Segawa, and Y. Ando, Phys. Rev. Lett. **93**, 107001 (2004).
[11] Y. Ando, S. Ono, X. F. Sun, J. Takeya, F. F. Balakirev, J. B. Betts, and G. S. Boebinger, Phys. Rev. Lett. **92**, 247004 (2004).
[12] M. Sutherland, S. Y. Li, D. G. Hawthorn, R. W. Hill, F. Ronning, M. A. Tanatar, J. Paglione, H. Zhang, L. Taillefer, J. DeBenedictis, R. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. Lett. **94**, 147004 (2005).
[13] Y. Ando, G. S. Boebinger, A. Passner, T. Kimura, and K. Kishio, Phys. Rev. Lett. **75**, 4662 (1995).
[14] G. S. Boebinger, Y. Ando, A. Passner, T. Kimura, M. Okuya, J. Shimoyama, K. Kishio, K. Tamasaku, N. Ichikawa, and S. Uchida, Phys. Rev. Lett. **77**, 5417 (1996).
[15] P. Fournier, P. Mohanty, E. Maiser, S. Darzens, T. Venkatesan, C. J. Lobb, G. Czjzek, R. A. Webb, and R. L. Greene, Phys. Rev. Lett. **81**, 4720 (1998).
[16] S. Ono, Y. Ando, T. Murayama, F. F. Balakirev, J. B. Betts, and G. S. Boebinger, Phys. Rev. Lett. **85**, 638 (2000).
[17] X. F. Sun, S. Komiya, J. Takeya, and Y. Ando, Phys. Rev. Lett. **90**, 117004 (2003); D. G. Hawthorn, R. W. Hill, C. Proust, F. Ronning, M. Sutherland, E. Boaknin, C. Lupien, M. A. Tanatar, J. Paglione, S. Wakimoto, H. Zhang, L. Taillefer, T. Kimura, M. Nohara, H. Takagi, and N. E. Hussey, *ibid.* **90**, 197004 (2003).
[18] K. Segawa and Y. Ando, Phys. Rev. Lett. **86**, 4907 (2001).
[19] K. Segawa and Y. Ando, Phys. Rev. B **69**, 104521 (2004).
[20] Y. Zhang, N. P. Ong, P. W. Anderson, D. A. Bonn, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **86**, 890 (2001).
[21] R. W. Hill, C. Lupien, M. Sutherland, E. Boaknin, D. G. Hawthorn, C. Proust, F. Ronning, L. Taillefer, R. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. Lett. **92**, 027001 (2004).
[22] Y. Hanaki, Y. Ando, S. Ono, and J. Takeya, Phys. Rev. B **64**, 172514 (2001).
[23] S. Komiya and Y. Ando, Phys. Rev. B **70**, 060503(R) (2004).
[24] Y. Ando, K. Segawa, S. Komiya, and A. N. Lavrov, Phys. Rev. Lett. **88**, 137005 (2002).
[25] S. A. Kivelson, I. P. Bindloss, E. Fradkin, V. Oganesyan, J. M. Tranquada, A. Kapitulnik, and C. Howald, Rev. Mod. Phys. **75**, 1201 (2003).
[26] S. Nakamae, K. Behnia, L. Balicas, F. Rullier-Albenque, H. Berger, and T. Tamegai, Phys. Rev. B **63**, 184509 (2001).
[27] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[28] The $\ln T$ behavior of weak localization should saturate at low enough T in a finite system, where the sample size sets the upper limit for L_i; in our data in Fig. 1(b), the slight saturation tendency discernible at the lowest T could be a similar finite-size effect, and it does not automatically suggest a conceptually metallic ground state.
[29] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt-Saunders, Philadelphia, 1976), p. 323.