Deviation from the Wiedemann-Franz law induced by nonmagnetic impurities in overdoped $La_{2-x}Sr_xCuO_4$

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(Received 5 June 2009; revised manuscript received 22 July 2009; published 25 September 2009)

To investigate the validity of the Wiedemann-Franz (WF) law in disordered but metallic cuprates, the low-temperature charge and heat transport properties are carefully studied for a series of impurity-substituted and carrier-overdoped La_{1.8}Sr_{0.2}Cu_{1-z} M_z O₄ (M=Zn or Mg) single crystals. With moderate impurity substitution concentrations of z=0.049 and 0.082 (M=Zn), the resistivity shows a clear metallic behavior at low temperature and the WF law is confirmed to be valid. With increasing impurity concentration to z=0.13 (M=Zn) or 0.15 (M=Mg), the resistivity shows a low-T upturn but its temperature dependence indicates a finite conductivity in the T→0 limit. In this weakly localized metallic state, that is intentionally achieved in the overdoped regime, a *negative* departure from the WF law is found, which is opposite to the theoretical expectation.

DOI: 10.1103/PhysRevB.80.104510

PACS number(s): 74.25.Fy, 74.72.Dn

I. INTRODUCTION

The Wiedemann-Franz (WF) law, which relates the thermal conductivity κ and the electrical conductivity σ through a simple formula,

 $\kappa/\sigma T = L,\tag{1}$

where L is called the Lorentz number and is given by the Sommerfeld's value $L_0 = 2.44 \times 10^{-8} \text{ W}\Omega/\text{K}^2$, is a robust signature of a Fermi liquid. It relies on the single-particle description of the transport properties and a purely elastic electron scattering. Although the WF law is usually not obeyed at $T \neq 0$ because of the importance of inelastic scattering, it must be obeyed in the Fermi liquid at T=0 where the electrons are elastically scattered by static disorders. The examination of the WF law at $T \rightarrow 0$ by using sub-Kelvin charge and heat transport measurements can therefore provide a direct judgment on the nature of the normal state of high- T_c cuprates, which is a prime example of the strongly correlated electron systems challenging the description of Fermi liquids. It has been found in the overdoped $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) that the WF law is perfectly obeyed, which indicated a Fermi-liquid ground state of the overdoped cuprates.¹

It was recently predicted that the WF law is violated in disordered interacting electron systems,^{2–4} in which the Coulomb interactions are likely to bring some additional scattering and impede the charge transport more efficiently. A recent study of the low-*T* transport of $\text{Bi}_{2+x}\text{Sr}_{2-x}\text{CuO}_{6+\delta}$ (Bi2201),⁵ in which a positive departure from the WF law $(\kappa/\sigma T > L_0)$ appeared at optimum doping and became more pronounced in the underdoped regime, was argued to support the theoretical prediction because the disorder in Bi2201 increases with decreasing doping level due to its peculiar chemistry. However, in previous studies, both on the over-

doped Tl2201 and on the underdoped Bi2201, the normal state was achieved by applying very strong magnetic field, which could induce some kind of spin-density/charge-density wave order.^{6–8} Such a competing order may cause some uncertainty regarding whether the properties of the true ground state is indeed detected.^{9,10} It would therefore be useful if one could study the validity of the WF law in cuprates whose normal states is obtained in a way other than by applying a strong magnetic field.

Introducing impurities or defects into CuO₂ planes is a viable way of suppressing the superconductivity and obtaining the normal state. Because of the non-*s*-wave symmetry of cuprates, nonmagnetic impurities such as Zn or Mg are found to destroy the superconductivity quite strongly, particularly for the underdoped cuprates.^{11–14} Furthermore, the impurity substitution was found to show different impacts on the physical properties in the underdoped and the overdoped cuprates, signifying a remarkable difference in the corresponding ground states.¹¹

It was found that in underdoped $La_{2-x}Sr_xCuO_4$ (LSCO) and $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$, a low-*T* resistivity divergence shows up immediately after the superconductivity is completely suppressed by a slight Zn substitution (~2%).^{11–13} The resistivity divergence is even stronger than the log(1/*T*) function, which is a well-known low-*T* behavior of high-magneticfield-induced insulating state,^{15,16} and therefore indicates an insulating ground state.^{12,13} The magnetoresistance data suggested that the Zn-induced insulating state is likely due to Kondo effect associated with impurity-induced magnetism.¹³ In any event, the normal state of these disordered and underdoped cuprates is apparently insulating and it is actually not meaningful to test the validity of the WF law for these samples.

In optimally doped or overdoped regimes, on the other hand, a slight Zn substitution ($\sim 4\%$) was found to induce a

simply metallic state when the superconductivity is suppressed,¹¹ which is believed to demonstrate the Fermiliquid ground state of the overdoped cuprates. However, it was also found that the heavy electron irradiation (to introduce point defects and to destroy the superconductivity similarly to Zn substitution) in the overdoped Tl2201 can lead to a low-T resistivity upturn,¹⁷ so the occurrence of electron localization in the overdoped regime appears to depend on the level of disorder. The temperature dependence of the conductivity suppression $\Delta\sigma$ (compared to the extrapolated residual conductivity from the high-T data) in the heavily electron-irradiated Tl2201 had a ln T variation and was in rough quantitative agreement with the two-dimensional (2D) weak localization theory. Once the Anderson transition into the insulating state takes place and the resistivity shows an exponentially divergent behavior due to strong disorder, even in the overdoped regime the WF law is meaningless. However, in the moderately disordered regime in which the superconductivity is suppressed by disorder but the resistivity is not really divergent, it becomes possible to examine the peculiar electron-transport properties through the WF law without introducing the possible complications from the magnetic-field-induced effects.

In this work, by substituting Zn or Mg for Cu in $La_{1.8}Sr_{0.2}Cu_{1-7}M_{7}O_{4}$ (M=Zn or Mg) (LSCMO), we are able to obtain a series of nonsuperconducting overdoped LSCO. When the superconductivity is just suppressed at Zn concentration of z=0.049 and 0.082, the ground state is clearly metallic and the WF law is found to hold reasonably well, pointing to the Fermi-liquid state of the overdoped cuprates. With higher concentration of Zn (z=0.13) or Mg (z=0.15), a low-T resistivity upturn shows up. It is found that the Tdependence of the conductivity obeys a $T^{1/2}$ law and therefore indicates a metallic ground state at $T \rightarrow 0$. This result, accompanied with the negative magnetoresistance, suggests a three-dimensional (3D) weak localization behavior. However, the low-T heat transport data indicate a surprisingly *negative* deviation from the WF law $(\kappa_0 / \sigma T < L_0)$ that is opposite to the theoretical prediction for the disordered interacting electron systems. This result demonstrates vet another unconventional feature of the mysterious normal state of the high- T_c cuprates.

II. EXPERIMENTS

High-quality $La_{1.8}Sr_{0.2}Cu_{1-z}M_zO_4$ (M=Zn or Mg) single crystals are grown by a traveling-solvent floating-zone method with similar processes to that reported in Ref. 18. The actual Zn or Mg concentration z is determined by the inductively coupled plasma atomic-emission spectroscopy. All the samples are carefully checked by using the x-ray Laue photograph and cut precisely along the crystallographic axes. The in-plane resistivity and the thermal conductivity are measured actually along the *a* axis. To remove the oxygen defects which tend to occur in overdoped LSCO, the crystals are annealed at 800 °C in sealed quartz tubes with 4 atm oxygen pressure for at least 5 days, followed by rapid quenching to the liquid nitrogen temperature. Several pieces of crystals used for transport measurements at each impurity



FIG. 1. (Color online) Temperature dependences of the in-plane resistivity of several Zn- and Mg-substituted LSCO single crystals. Two pieces of samples (labeled A and B) for each impurity concentration are measured for checking the uncertainties of the transport data. The small discrepancy (<10%) of the resistivity data between different samples at the same impurity concentration indicates a high quality of the crystals.

concentration are labeled "A," "B," "C," etc. The in-plane resistivity ρ_{ab} is measured using a standard ac four-probe method in a ⁴He cryostat from 1.4 to 300 K and using a LR-700 ac-resistance bridge in a dilution refrigerator from 80 to 900 mK, respectively. In the latter case, excitation voltages are minimized to avoid observable self-heating effect. The thermal conductivity measurement in the sub-Kelvin region is done by a conventional steady-state "one heater, two thermometer" technique with RuO₂ sensors in the dilution refrigerator.¹⁹

III. RESULTS AND DISCUSSION

A. Low-temperature resistivity

Figure 1 shows the in-plane resistivity ρ_{ab} of several Znand Mg-substituted LSCO single crystals with Sr content x =0.2. The pristine LSCO sample shows a good T-linear behavior of ρ_{ab} below 300 K and a sharp superconducting transition at 34.5 K. With increasing Zn concentration to z =0.049 and 0.082, the linearity of the resistivity becomes a bit worse but obviously a metallic behavior (with positive temperature coefficient) persists down to very low temperature without displaying any superconducting transition. When Zn concentration increases to 0.13, the resistivity starts to show a weak upturn below ~ 12 K. In Mgsubstituted samples (z=0.15), the low-temperature upturn of the resistivity is pronounced. These data demonstrate the impact of impurity or disorder on the electron transport and superconductivity, all consistent with the literature.²⁰ In particular, the low-T resistivity upturn in the heavily Zn- or Mg-substituted samples are qualitatively the same as that in the electron irradiated Tl2201.¹⁷

To further investigate the low-T transport properties of highly Zn- and Mg-substituted LSCO, the in-plane resistivity is studied down to milli-Kelvin temperature and in magnetic



FIG. 2. (Color online) [(a) and (b)] Low-temperature resistivity of Zn-substituted (z=0.13) and Mg-substituted (z=0.15) LSCO single crystals in different magnetic fields (0–16 T and along the *c* axis). [(c) and (d)] The electrical conductivity of the above two samples in 0 and 16 T fields, plotted in σ_{ab} vs $T^{1/2}$. The solid lines are linear fittings to the low-temperature data.

fields from 0 to 16 T ($||c\rangle$). The detailed data are shown in Figs. 2(a) and 2(b) in log *T* plots. First of all, the resistivity of these samples keeps increasing upon lowering temperature down to the lowest temperature (~80 mK). However, the increase in resistivity is apparently weaker than the log(1/*T*) function, which is a common insulating behavior in both the high-magnetic-field-induced normal state of underdoped high- T_c cuprates and the lightly doped nonsuperconducting YBa₂Cu₃O_y (YBCO).^{15,16,21} This observation suggests that the resistivity may not diverge in the zero-temperature limit in these samples. It is actually easy to find, as shown in Figs. 2(c) and 2(d), that the low-*T* electrical conductivity of these samples follows a power law

$$\sigma_{ab} = \sigma_0 + \beta T^{1/2}, \qquad (2)$$

where σ_0 and β are temperature-independent coefficients. It follows from this temperature dependence that the resistivity $1/(\sigma_0 + \beta T^{1/2})$ remains finite at $T \rightarrow 0$, which is a clear indication of a metallic ground state of these samples. Note that this T dependence of resistivity is somehow different from the ln T variation of $\Delta \sigma$ observed in the overdoped Tl2201 after significant electron irradiation.¹⁷ Since the measurements of Tl2201 were not done at very low temperatures, a direct comparison between LSCMO and Tl2201 is not available now. Nevertheless, the 2D weak localization picture proposed for Tl2201, which is based on the ln T behavior of conductivity, may not be appropriate for LSCMO. Remember, the particular $T^{1/2}$ dependence of conductivity is known for the 3D weak localization.^{22,23} It is possible that the overdoped LSCO with strong disorder is effectively a 3D electron system, though the temperature dependence of the resistivity anisotropy suggests that clean LSCO is two dimensional for the whole superconducting compositions (x < 0.30).²⁴

Another important feature one may note in Fig. 2 is that the low-*T* resistivity decreases in magnetic fields, that is, they show rather strong negative magnetoresistance. This immediately rules out the possible contamination of the superconducting fluctuations to the resistivity data and suggests that the weak negative temperature coefficient of resistivity is due to the intrinsic electron transport of the normal state. Furthermore, the negative magnetoresistance at low temperature also indicates that the weak localization is likely responsible for the resistivity upturn.^{22,23}

As a summary of the electrical transport results, the resistivity upturn in the heavily Zn and Mg substituted and carrier-overdoped LSCO does not signify a metal-toinsulator crossover (MIC) but is attributed to an inelastic electron scattering by impurities/disorders in a metallic regime. Furthermore, the temperature dependence of the low-*T* resistivity does not suggest a Kondo-type scattering mechanism, which is often playing a role in the charge transport in underdoped cuprates.^{13,25}

B. Low-temperature thermal conductivity of simply metallic samples

To further investigate the electron-transport properties, the thermal conductivity κ of Zn- and Mg-substituted LSCO samples shown in Fig. 1 have been measured down to 70 mK; Fig. 3 shows the data for the superconducting La_{1.8}Sr_{0.2}CuO₄ sample and LSCMO samples with Zn concentration of 0.049 and 0.082, which showed the metallic resistivity behavior down to very low temperatures. Note that the low-*T* thermal conductivity of all these samples can be well fitted either to

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + bT^2,\tag{3}$$

or to

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + AT,\tag{4}$$

as shown in Figs. 3(a) and 3(b), respectively, but the temperature range for obtaining a good fit is much wider with using Eq. (4). Looking at Fig. 3, one may notice that the data quality of these charge-carrier-overdoped LSCO samples are unprecedentedly high. In a previous study,²⁶ some clear downturn of the low-*T* thermal conductivity data was observed in the overdoped LSCO and was already discussed to be due to the electron-phonon decoupling,²⁷ whose adverse effects on probing the intrinsic electron thermal conductivity can be avoided if the contacts are good enough.

Note that Eq. (3) is commonly used for describing the low-*T* thermal conductivity of superconducting cuprates, where the residual term κ_0/T is the contribution of *d*-wave nodal quasiparticles and bT^2 is the phonon contribution in the boundary scattering limit.^{28–31} In this regard, the present data of the pristine LSCO are in good harmony with the



FIG. 3. (Color online) Low-temperature thermal conductivity of Zn-substituted LSCO single crystals, whose resistivity data are shown in Fig. 1. The data are plotted in (a) κ/T vs T^2 and (b) κ/T vs T. The solid lines are linear fittings to the low-temperature data in two different plots. Note the difference in the residual term obtained by the two kinds of analysis. The solid symbols at T=0 K indicate the values of κ/T calculated by using the Wiedemann-Franz law with the residual resistivity obtained from Fig. 1. Note that the calculated κ_0/T and the experimental κ/T data are shown with the same shape of the symbols for the same samples.

previous studies. For samples with Zn concentrations of 0.049 and 0.082, whose ground state is metallic as indicated by the in-plane resistivity, κ_0/T is apparently coming from the electron heat transport as in ordinary metals. If the ground state is a Fermi liquid, one can expect that the values of the residual resistivity and κ_0/T satisfy the WF law. From Fig. 3(a), it is clear that there is about 20% difference between the κ_0/T obtained from Eq. (3) and the values calculated from the residual resistivity (see Fig. 1) using the WF law. Note that this 20% difference is not mainly due to the experimental error, since the resistivity and thermal conductivity measurements are performed using the same samples and the same contacts.

It was recently reported that Eq. (4) is a good description for the low-*T* thermal conductivity of Tl2201 and Bi₂Sr₂CaCu₂O_{8+ δ}.^{19,32} As shown in Fig. 3(b), fitting the present thermal conductivity data to Eq. (4) gives qualitatively the same behavior upon Zn substitution as that in Fig. 3(a), but the κ_0/T from this fitting agree better (in fact, almost perfectly) with those from the WF law. This gives a circumstantial support to the validity of Eq. (4) in the overdoped regime.

Compared to the underdoped LSCO, in which a modified formula with the phonon boundary scattering $\kappa/T = \kappa_0/T$ $+b'T^{\alpha-1}$ ($\alpha=2.5-3$) was proposed to fit the data well,²⁶ the second term AT in Eq. (4) is apparently not originating from the boundary scattering limit. One possible explanation for this unusual power is the effect of strong electron-phonon scattering on phonons. It has been known that a T^2 behavior of the phonon thermal conductivity can show up in both usual metals and *d*-wave superconductors.^{33,34} In the case of a *d*-wave superconductor, the Fermi surface is anisotropicaly gapped except for the gap nodes, nearby where the quasiparticles are excited even at low temperature.^{35,36} Those socalled nodal quasiparticles dominate the transport properties.³⁶ For such a system, it was theoretically proposed that the T^2 behavior of the phonon thermal conductivity originates from the fact that the transverse phonons propagating along certain high-symmetry directions do not interact with the nodal quasiparticles.³⁴ However, the nonsuperconducting overdoped samples have a complete Fermi surface, and therefore a very different momentum distribution of electrons from that in *d*-wave superconductors is formed. Apparently, the electron-phonon scattering should differ significantly for the Zn-free and Zn-substituted samples, considering the distinct difference in the electronic structures between the superconducting and nonsuperconducting samples. Therefore, the T^2 coefficient is expected to be quite different in these two cases. Nevertheless, as shown in Fig. 3, the coefficient A of the pristine (superconducting) and Znsubstituted (nonsuperconducting) LSCO are found to be nearly the same, which seems to rule out the possibility that the T^2 law comes from the electron-phonon scattering. In addition, one may notice that, when the electron-phonon scattering is strong, the electronic thermal conductivity κ_{e}/T cannot be temperature independent and therefore Eq. (4) is physically imprecise.32

Another possible origin of the AT term is the phonon thermal conductivity with the dominant scattering of dislocations.³³ This scenario is consistent with the observation that Eq. (3) becomes a good description of the data at very low temperatures, because the dislocation scattering is likely to be smeared out at low enough temperatures. There is a possibility that some dislocation defects are produced in LSCO crystals when they are quenched to nitrogen temperature after high-temperature annealing.

In any case, it appears that the WF law holds reasonably well (the deviation is no more than 20%) in the overdoped LSCO when the normal state is obtained by a moderate impurity substitution. If one could take the validity of the WF law as a robust signature of a Fermi-liquid state, the present result apparently indicates a Fermi-liquid nature of the overdoped LSCO. This conclusion is essentially the same as that for the overdoped Tl2201, where the normal state was achieved by strong magnetic fields.¹ However, one should note that the resistivity of our moderately Zn-substituted samples actually shows a good *T*-linear behavior at low tem-



FIG. 4. (Color online) Low-temperature thermal conductivity of Zn-substituted (z=0.13) and Mg-substituted (z=0.15) LSCO single crystals. (a) and (b) data are plotted in κ/T vs T^2 and the linear relationship is not observed even in the lowest temperature regime. (c) and (d) data are plotted in κ/T vs T and κ/T vs $T^{0.8}$ for Zn- and Mg-substituted samples, respectively. Linear fittings to the low-temperature data are shown by solid lines. The arrows indicate the residual thermal conductivity κ_0/T calculated by using the Wiedemann-Franz law from the residual electrical conductivity σ_0 of the data shown in Fig. 2; note that they are very different from the extrapolated residual values indicated by the linear fitting to the experimental data.

perature, which has been taken as a characteristic non-Fermiliquid feature.³⁷ Therefore, there is still some inconsistency between the interpretations of the WF law and the resistivity behavior in overdoped LSCO, which needs further clarification.

C. Low-temperature thermal conductivity for samples showing resistivity upturn

The low-temperature thermal conductivity data of LSCMO with higher impurity concentration z=0.13 (M=Zn) and 0.15 (M=Mg) are shown in Fig. 4. From Figs. 4(a) and 4(b), it can be seen that the usual formula of Eq. (3) is not able to fit the low-*T* thermal conductivity data, that is, the experimental data plotted in κ/T vs T^2 never show a linear behavior down to ~60 mK; in fact, the curves always have a negative curvature. This phenomenon is very similar to what we observed in the lightly doped nonsuperconducting YBCO,²¹ in which case the heading of κ/T to zero upon lowering temperatures and the log(1/*T*)-divergent resistivity demonstrated the insulating ground state.

In the present case, we would like to point out at the beginning that the curvature of κ/T vs T^2 plots is *not* likely due to the decoupling of electrons and phonons.^{26,27} It is known that the necessity of making good contacts to obtain the intrinsic electronic thermal conductivity data becomes

more severe when the electron term becomes larger. Comparing the data shown in Figs. 3 and 4, one can easily see that the pristine and lower Zn-substituted LSCO samples, which must have larger electron thermal conductivity than z=0.13 (M=Zn) and 0.15 (M=Mg) samples, actually do not show obvious sign of downturn curvature in their κ/T vs T^2 plots.

Upon further analyzing the data, one can find that Eq. (4) is still able to fit the thermal conductivity data of z=0.13 (M=Zn) samples very well from 60 to 400 mK, as shown in Fig. 4(c). Because of the even stronger curvature in the κ/T data of z=0.15 (M=Mg) sample, neither Eq. (3) nor Eq. (4) can fit the data; instead, the low-T data of Mg-substituted LSCO can be described by a formula

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + A' T^{0.8},\tag{5}$$

with the fitting parameter $\kappa_0/T=0$.

As we have seen in Sec. III A, the charge transport data indicate that in the heavily Zn- or Mg-substituted LSCO some inelastic scattering is taking place at low temperatures, causing a weak localization behavior. One direct consequence of this fact is that κ_e/T is no longer independent of temperature in these samples, and the resistivity upturn naturally corresponds to a gradual decrease of κ_e/T with lowering temperatures. Obviously, this invalidates Eq. (4). Neverthe less, one can apparently fit the data for z=0.13 (M=Zn) samples to Eq. (4), which is probably due to the fact that the low-T resistivity upturn (<10% increase in ρ_{ab} from 900 to 80 mK) or the inelastic scattering are weak enough to make the deviations of κ_e/T from a constant effectively negligible. In contrast, the z=0.15 (M=Mg) samples show even stronger resistivity upturn and more significant inelastic scattering. It is likely that a considerable temperature dependence of the electronic term κ_e/T is the cause of Eq. (5), which is an approximate description of the low-T thermal conductivity of Mg-substituted sample.

Based on the linear fittings (shown in Fig. 4) to obtain the residual term κ_0/T , one can easily find an important result, that is, the WF law in these metallic samples are strongly violated. As shown in Figs. 4(c) and 4(d), the κ_0/T values obtained from the extrapolation of the thermal conductivity data to T=0 K are much smaller than those predicted by the WF law using the residual conductivity shown in Figs. 2(c)and 2(d). Especially, the Mg-substituted LSCO crystal gives a κ_0/T value that is essentially zero, indicating a "thermal insulator" ground state, while the resistivity data suggest a finite residual term and a metallic ground state. It is surprising that the departure from the WF law is negative, which has never been observed in cuprates; for example, upon approaching MIC in the underdoped region of cuprates, the departure from the WF law was found to be positive.⁵ Intriguingly, there is another metallic system which exhibits a negative deviation from the WF law: It is the heavy-fermion system CeCoIn₅ when tuned to its quantum critical point (QCP).³⁸ In that materials, the violation of the WF law is accompanied by a T-linear resistivity and has been discussed to be due to an anisotropic destruction of the Fermi surface. Such a QCP-related origin of the WF-law violation is unlikely for LSCO, given that the violation is not observed in less disordered (z=0.049 and 0.082) samples. In passing, it is useful to note that the negative departure from the WF law is contrary to the theoretical predictions for the disordered interacting electron systems.^{2–4} Although one could speculate that this unusual breakdown of the WF law in LSCO may be related to the strong electron correlations or the antiferromagnetic spin fluctuations in cuprates, the validity of the WF law in samples with lower Zn concentrations does not support this speculation. Hence, the charge and heat transport properties in the moderately disordered, weakly localized regime call for deeper theoretical investigations on systems in the vicinity of electron localization.

IV. SUMMARY

Both the charge and heat transports properties are studied in overdoped LSCO where the superconductivity is completely suppressed by Zn or Mg substitution. When the superconductivity is just destroyed with the Zn substitution of

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z=0.049 and 0.082, the ground state is clearly metallic and the WF law is found to hold reasonably well, pointing to the Fermi-liquid nature of the overdoped cuprates. With higher doping of Zn (z=0.13) or Mg (z=0.15), a low-T resistivity upturn shows up but the T dependence of the electrical conductivity follows a $T^{1/2}$ law and therefore indicates a metallic ground state. In this metallic state, which is in the regime of 3D weak localization, the low-T transport data indicate a surprisingly negative deviation from the WF law that is opposite to the theoretical prediction for the disordered interacting electron systems.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Grants No. 10774137 and No. 50721061), the National Basic Research Program of China (Grants No. 2006CB922005 and No. 2009CB929502), the Research Fund for the Doctoral Program of Higher Education of China (Grant No. 20070358076), and KAKENHI Grants No. 19674002 and No. 20030004.

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