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Electronic ground state of heavily overdoped nonsuperconducting $La_{2-x}Sr_xCuO_4$

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We report detailed thermodynamic and transport measurements for nonsuperconducting $La_{1.7}Sr_{0.3}CuO_4$. Collectively, these data support the presence of a highly correlated Fermi-liquid ground state in $La_{2-x}Sr_xCuO_4$ beyond the superconducting dome, and imply that charge transport in the cuprates is dominated at finite temperatures by electron-electron scattering.

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The high-T_c cuprates (HTC's) have emerged as one of the most formidable challenges in solid-state physics.[1] In particular, transport properties of the normal state present a number of uneasy paradoxes for the Fermi-liquid (FL) picture. While it has been argued that strong electron-phonon (e-ph) scattering might account for the T-linear resistivity in optimally doped cuprates,[2] it is generally assumed that proximity to a Mott insulator, and thus the unusual strength of on-site electronic repulsion, is the fundamental reason behind the more unusual aspects of HTC physics. This general assumption is backed up by a host of experimental observations which indicate that the anomalous behavior becomes even more pronounced with decreasing density of carriers, that is, by moving towards the underdoped side. At sufficiently high carrier concentrations however, it has often been assumed that HTC's eventually evolve into a conventional FL as the electron correlations become weaker and the system becomes more three dimensional (3D).

Ironically, the persistence of robust superconductivity on the overdoped (OD) side of the phase diagram has been a major obstacle in the exploration of the metallic nonsuperconducting ground state in HTC's. Indeed, supporting evidence for a FL ground state has only surfaced very recently with the experimental verification of the Wiedemann-Franz (WF) law in OD $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) ($T_c \sim 15$ K).[3] By suppressing superconductivity in a large magnetic field, Proust *et al.* observed the precise WF ratio $\kappa_{ab}/\sigma_{ab}T = L_0$, where κ_{ab} and σ_{ab} are the in-plane thermal and electrical conductivities and the Lorenz number $L_0 = 2.44 \times 10^{-8}$ $\mathrm{W}\Omega/\mathrm{K}^{-2}$. Surprisingly however, and at odds with a conventional FL picture, the WF relation was found to coexist with a large linear resistivity term extending down to 0 K. This dichotomy raises the question whether the field-induced "normal state" in OD HTC's, i.e., beyond H_{c2}, is the same as the ground state that would exist in the absence of a magnetic field, as it does in more conventional superconductors. Moreover, a clear understanding of the experimental situation in OD cuprates has often been compounded by their tendency to undergo phase separation.

In this communication, we present in- (ρ_{ab}) and out-of-plane (ρ_c) resistivity, in- (κ_{ab}) and out-of-plane (κ_c) thermal conductivity, specific heat (C) and magnetic susceptibility (χ) measurements on single-phase nonsuperconducting $La_{2-x}Sr_xCuO_4$ (LSCO) crystals ($x = 0.30$) in which these various concerns are removed. Collectively, the data provide a consistent picture of $La_{1.7}Sr_{0.3}CuO_4$ as a highly correlated FL. The WF law is verified to within our experimental resolution, though in contrast to OD Tl2201, both ρ_{ab} and ρ_c exhibit strictly T^2 behavior below 50 K, *with no additional linear term*. Significantly, the Kadowaki-Woods ratio, linking the coefficient A of the (in-plane) T^2 resistivity and the square of the linear specific-heat coefficient γ_0, is found to be anomalously enhanced, even compared with other strongly correlated metals. This latter observation implies that intense e-e scattering persists beyond the superconducting dome and sheds more light on the evolution of $\rho_{ab}(T)$ across the HTC phase diagram.

Seven bar-shaped samples (typical dimensions 3 mm $\times 0.6$ mm$\times 0.6$ mm) were prepared for either ab-plane (A1-4) or c-axis (C1-3) measurements from a large $La_{1.7}Sr_{0.3}CuO_4$ single crystal grown in an infrared image furnace. The individual ingots were post-annealed together with the remaining boule under extremely high partial pressures of oxygen (400 atm) for 2 weeks at 900 °C to minimize oxygen deficiencies and to ensure good homogeneity within each crystal. Subsequent x-ray analysis revealed good crystallinity and no trace of superconductivity could be detected (resistively) down to 95 mK (see top inset to Fig. 1), confirming that these crystals were indeed single phase. $\chi(T)$ of the boule was measured with a commercial magnetometer, while $C(T)$ was measured between 0.6 K and 10 K in a relaxation calorimeter. In both cases, addenda contributions were measured independently and subtracted from the raw data. $\kappa(T)$ of $A1$, $A2$, and $C1$ were measured in a dilution refrigerator using three RuO_2 chips employed as one heater and two thermometers. $\rho(T)$ measurements were made on all seven samples using a conventional ac four-probe method. For all transport measurements, current and voltage contacts were painted onto the crystals so as to short out any spurious voltage drops from orthogonal components of the conductivity tensor. Uncertainty in the geometrical factor was esti-

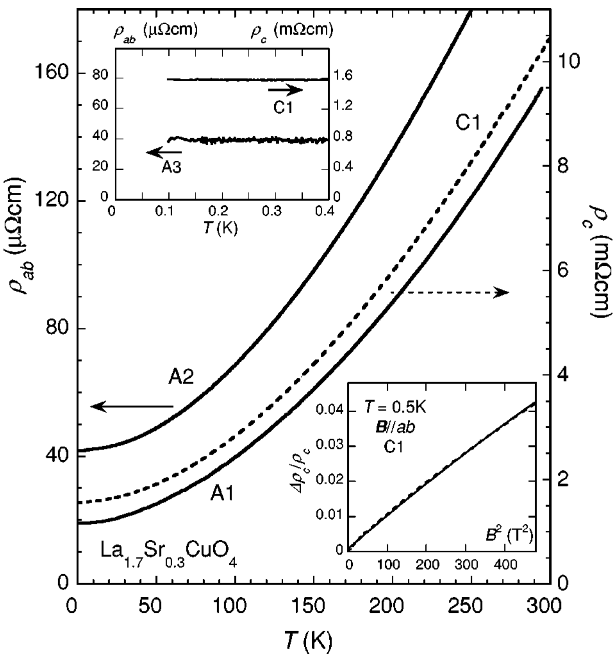

FIG. 1. Zero-field resistivity of $A1$, $A2$ (ρ_{ab}, solid lines), and $C1$ (ρ_c dashed line). Upper inset: Low-T $\rho(T)$ of $A3$ and $C1$. Lower inset: $\Delta\rho_c/\rho_c$ vs B^2 for $C1$ at 0.5 K ($B\|ab$). The solid line is a fit to $\Delta\rho_c/\rho_c = 0.000\,10B^2 - 3.1\times10^{-8}B^4$.

mated to be $\sim$20%. Finally, c-axis magnetoresistance $\Delta\rho_c/\rho_c$ data were taken on $C1$ at the NHMFL in Florida.

Figure 1 shows $\rho(T)$ of $A1$, $A2$, and $C1$ below 300 K. (All crystals reported here showed similar behavior.) Note the similar metallic T dependencies observed along both orthogonal directions, the different residual resistivities (ρ_0) for $A1$ and $A2$, and the strong upward curvature across the entire temperature range. The resistivity ratio $\rho_c(C1)/\rho_{ab}(A1)$ rises from $\sim$65 at 300 K to $\sim$80 as $T\to 0$, presumably due to slight differences in their ρ_0 values. The limiting low-T resistivities of $C1$ and $A3$ are reproduced in the top inset to Fig. 1. No current dependence was observed in $\rho(T)$ down to 0.1 A/cm^2, well below typical critical current densities found in HTC. The lower inset shows $\Delta\rho_c/\rho_c$ for $C1$ ($B\|ab$) plotted versus B^2 at $T=0.5$ K. Inserting the fit to the low-field data (see caption) into the Boltzmann transport equation for a quasi-2D FL (see, e.g., Ref. 4), we obtain an estimate of the in-plane electronic mean free path $\ell_{ab}\sim 145$ Å. Finally, using $k_F=0.55$ Å^{-1} for $La_{1.7}Sr_{0.3}CuO_4$ (Ref. 5) and the "isotropic-ℓ" approximation,[6] we obtain $\rho_{ab0}=21\ \mu\Omega$ cm. This value is in good agreement with $\rho_0(A1)$ and implies that *all* carriers contribute to the metallic conductivity.

In the top panel of Fig. 2, $\rho(T)$ of $A3$, $A4$, and $C2$ are plotted versus $T^{1.6}$. In a previous study,[7] such noninteger power-law resistivities were shown to extend from the lowest temperatures up to 1000 K. While $\rho(T)$ of our crystals follows very closely a $T^{1.6}$ dependence at elevated temperatures, the T dependence clearly becomes stronger than $T^{1.6}$ below $T\approx 100$ K. Indeed, as shown in the bottom panel in Fig. 2, $(\rho\text{-}\rho_0)(T)=AT^2$ up to at least $T=55$ K *for both current directions* [$(\rho-\rho_0)(T)$ for $A4$ is shown in an inset for

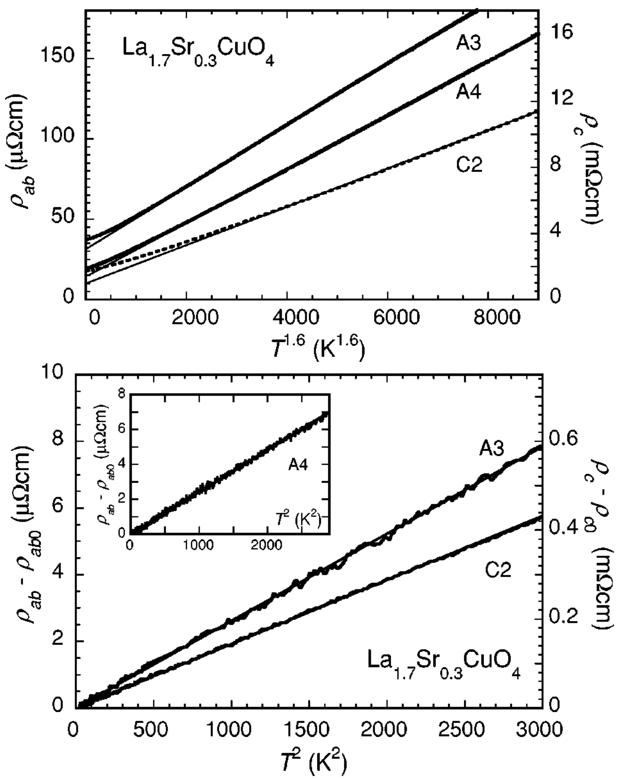

FIG. 2. (a) $\rho(T)$ of $A3$, $A4$ (solid lines), and $C2$ (dashed line) vs $T^{1.6}$. Thin solid lines are provided as guides. (b) Low-T $\rho(T)$ of $A3$ and $C2$ vs T^2. Inset: ρ vs T^2 for $A4$.

clarity]. Significantly, A has the same magnitude ($\sim 2.5 \pm 0.1\ n\Omega$ cm/K^2) for all in-plane crystals, even though $\rho_0(A2,A3)\sim 2\rho_0(A1,A4)$. To our knowledge, this is the first time a pure T^2 resistivity has been resolved in a hole-doped cuprate [recall that in OD Tl2201, $(\rho-\rho_0)(T)=\alpha T+AT^2$ (Ref. 3)] and coupled with good Matthiessen's rule behavior implies that in $La_{1.7}Sr_{0.3}CuO_4$ at least, charge transport is consistent with an anisotropic 3D FL picture.[8]

In Fig. 3, the low-T thermal conductivities of $A1$, $A2$, and $C1$ are plotted as κ/T versus T^2. Note that $A1$ and $A2$ are better conductors of heat, as expected since $\sigma_c \ll \sigma_{ab}$, and that $\kappa(A1)>\kappa(A2)$, reflecting their respective ρ_0 values. The sizable phonon term κ_{ph} in HTC's has often been a major obstacle to the determination of κ_{el}, the electronic contribution to $\kappa(T)$. In this instance, however, we can make use of the fact that $A1$ and $A2$ have different residual resistivities and simply compare the difference in the residual *conductivities* $\Delta\sigma_0$ with the difference in κ_{ab} for the two crystals. This comparison is shown in the left-hand-side inset to Fig. 3. Between 0.3 K and 1 K, $\Delta\kappa_{ab}/T\approx 0.57$ mW/cm K^2 (the dotted line). This compares favorably with our estimate (dashed line) of $\Delta\kappa_{ab}/T=L_0/\Delta\sigma_0=0.70$ mW/cm K^2. A similar conclusion can be deduced by comparing κ_{ab} with $\kappa_c(C1)$, if one assumes that κ_{ph} is anisotropic and introduce a scaling factor f (in this case, $f=1.6$) between the in-plane and out-of-plane κ_{ph}, i.e., $\kappa_{el}=\kappa_{ab}-1.6\kappa_c$. (Note that we do not expect anisotropy in κ_{ph} to be temperature dependent below 1 K.) The resultant κ_{el}/T, shown in the second inset to Fig. 3, are found to be 1.07 and 0.50 mW/

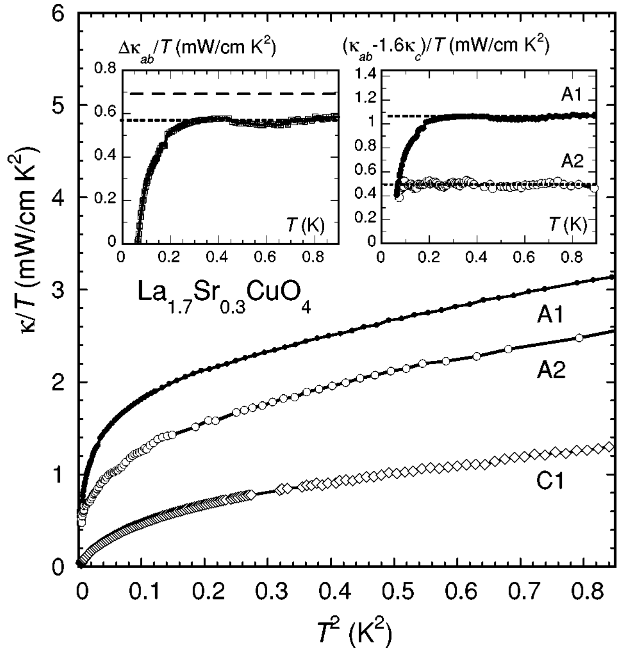

FIG. 3. Thermal conductivity κ/T of $A1$, $A2$, and $C1$ vs T^2. Left inset: $\Delta\kappa_{ab}/T$ vs T. The dashed line gives the expected $\Delta\kappa_{ab}/T = L_0/\Delta\sigma_0$ obtained from the differences in the residual resistivities. Right inset: $(\kappa_{ab} - 1.6\kappa_c)/T$ for $A1$ and $A2$. The dotted lines give κ_{el}/T as $T \rightarrow 0$.

cm K^2 for $A1$ and $A2$, respectively (the dotted lines). This gives $\rho_0 = L_0 T/\kappa_{el} = 23$ ($A1$) and 49 ($A2$) $\mu\Omega$ cm, compared with the measured values of 19 and 42 $\mu\Omega$ cm. Given our experimental uncertainty, the WF law appears to be well satisfied in both our in-plane $La_{1.7}Sr_{0.3}CuO_4$ crystals, at least down to 0.3 K. It should be stressed here that σ_{ab0}(LSCO) is almost an order of magnitude smaller than in OD Tl2201,[3] thus making separation of κ_{el} from κ_{ab} more difficult.

The surprising feature of Fig. 3 is of course the strong downturn in $\kappa(A1)$ as $T \rightarrow 0$. (A slight downturn is also visible in the second inset for $A2$ below 0.1 K, though nowhere near as dramatic.) Note that this downturn is robust both to changes in the thermal contacts and the crystal dimensions. Intriguingly, a vanishing of κ_{el} has also been observed in the electron-doped cuprate $Pr_{2-x}Ce_xCuO_4$.[9] In the normal state above H_{c2}, Hill *et al.* found that κ_{el}/T, while violating the WF law for $T > 0.3$ K (in the sense that it clearly exceeds L_0/ρ), rapidly vanishes below 0.3 K. These two anomalous observations appear to have a common origin. The sample-dependent aspect of the downturn, however, suggests that it is extrinsic and unrelated to any exotic electronic behavior, particularly in view of the other results obtained here for $La_{1.7}Sr_{0.3}CuO_4$ above 0.3 K. We reserve a detailed discussion on the origin of this anomalous downturn to a more complete and systematic investigation.[10]

The absence of superconductivity in $La_{1.7}Sr_{0.3}CuO_4$, coupled with the emergence of a T^2 resistivity at low T, gives us a unique opportunity to compare experimental manifestations of e-e correlations in $La_{1.7}Sr_{0.3}CuO_4$ to other strongly correlated systems. Electronic correlations in a FL are known to lead to an enhancement in the quasiparticle

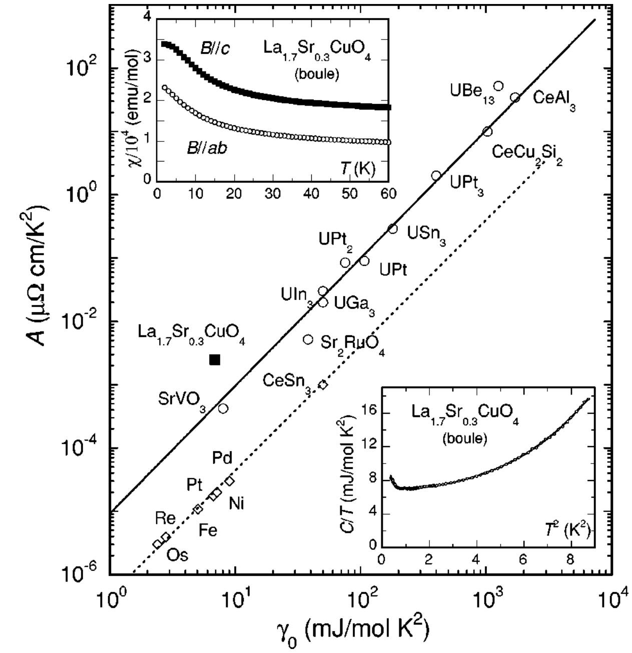

FIG. 4. Kadowaki-Woods plot of A, the coefficient of the T^2 resistivity, vs γ_0, the electronic specific-heat coefficient, for a variety of strongly correlated metals (adapted from Refs. 12, 13, and 16). Upper inset: Magnetic susceptibility $\chi(T)$ of large boule at 5 T with $B \| c$ (solid squares) and $B \| ab$ (open circles). Lower inset: Low-T specific heat of boule plotted as C/T vs T^2. The solid line is a fit to the expression $C = \alpha T^{-2} + \gamma_0 T + \beta_3 T^3 + \beta_5 T^5$. See text for parameter values.

effective mass. This effect can be detected through the resistivity (AT^2), specific heat ($\gamma_0 T$), and magnetic susceptibility (χ_p). Empirical relationships that correlate these physical parameters have been found in a wide range of strongly correlated metals; namely, the Kadowaki-Woods (KW) ratio $[A/\gamma_0^2 = a_0 = 10^{-5} \mu\Omega\ \mathrm{cm/(mJ/K\,mol)}^2$ (Refs. 11 and 12)] and the Wilson ratio $[R_w = (\pi k_B^2/3\mu_B^2)\chi_p/\gamma_0$, with $R_w \sim 1$ for a free-electron gas and ~ 2 for strongly correlated fermions[13]].

The lower inset to Fig. 4 shows $C(T)$ of the large crystal boule plotted as C/T versus T^2. $C(T)$ can be fitted over the whole temperature range ($0.6\ \mathrm{K} < T < 10\ \mathrm{K}$) to the expression $C = \alpha T^{-2} + \gamma_0 T + \beta_3 T^3 + \beta_5 T^5$ with $\alpha = 76\ \mu\mathrm{J\,K/mol}$, $\gamma_0 = 6.9\ \mathrm{mJ/mol\,K^2}$, $\beta_3 = 0.1\ \mathrm{mJ/mol\,K^4}$, and $\beta_5 = 0.7\ \mu\mathrm{J/mol\,K^6}$. The αT^{-2} term represents the high-temperature tail of a Schottky anomaly that invariably develops in HTC samples at low T due to either a small concentration of isolated paramagnetic impurities, or due to nuclear hyperfine or quadrupole splitting. The phononic $\beta_3 T^3$ term gives rise to a rather elevated $\Theta_D = 550$ K, though β_3 here may be made artificially low by a T-dependent electronic term.[14] The magnitude of γ_0, however, is very robust in our measurements,[15] is comparable to previous reports at this doping level,[14] and gives a corresponding KW ratio $A/\gamma_0^2 \sim 5a_0$. This puts nonsuperconducting LSCO well off the so-called "universal" line for strongly correlated metals indicated in Fig. 4. A similarly enhanced KW ratio, i.e., $A/\gamma_0^2 \geqslant 5a_0$, can also be inferred for OD Tl2201

[$A=5.4$ nΩ cm K^{-2} (Ref. 3), $\gamma_0 \sim 10$ mJ/mol K^2 (Ref. 17)], and indirectly for OD $Pr_{2-x}Ce_xCuO_4$, [$A \sim 4$ nΩ cm K^{-2} (Ref. 18), $\gamma_0 \sim 6.7$ mJ/mol K^2 (Ref. 19)]. This observation reveals an important aspect of the physics of the cuprates and is the central result of this Communication. While the origin of this enhanced KW ratio is not understood at present, it implies that in HTC's, correlation effects lead to an enhancement in the *e*-*e* scattering intensity without an accompanying change in the density of states. Clearly further investigations to establish whether this a common feature of *all* the cuprate families are strongly recommended.

As one moves across the HTC phase diagram towards half filling, one expects *e*-*e* scattering to become even more intense. Thus, the gradual evolution from quadratic to linear resistivity in HTC's may simply reflect the growing strength of *e*-*e* interactions as one approaches the Mott insulator from the metallic side. As an indication of how rapidly the *e*-*e* scattering intensity might grow, we note here that ρ_{ab} of $La_{1.85}Sr_{0.15}CuO_4$ is approximately two to three times larger than that of $La_{1.7}Sr_{0.3}CuO_4$ at 300 K, even though hole counting dictates that their carrier densities differ by only a few percent. Whilst the strength of the *e*-ph interaction may change across the phase diagram,[20] given that *e*-ph scattering appears to give a negligible contribution to $\rho_{ab}(T)$ in $La_{1.7}Sr_{0.3}CuO_4$, an *e*-ph origin of the linear resistivity that appears (in a narrow composition range) around optimal doping would appear unlikely. In light of these results, and in the absence of any other known mechanism, we can only conclude that chronic (Umklapp) *e*-*e* scattering processes dominate the normal-state transport behavior of the cuprates across the *entire* accessible doping range.[21] Such a scenario is consistent with the large increase in the quasiparticle lifetime observed below T_c.[22,23]

Finally, let us comment briefly on the Wilson ratio R_W. As shown in the upper inset to Fig. 4, $\chi(T)$ displays significant anisotropy with respect to the field orientation (believed to arise from anisotropic g values[24]) and a strong enhancement at low T. Assuming a constant Pauli susceptibility[25] χ_p and an isotropic Curie-Weiss term (with $\Theta_C \sim 7.5$ K), we obtain an average $\chi_p \sim 1.75\times10^{-4}$ emu/mol and hence, $R_W \sim 2.0$, consistent with values for other strongly correlated metals.[13] If however, all the enhancement in $\chi(T)$ is assumed to be intrinsic, i.e., due to an enhanced $\chi_p(T)$, we obtain $R_W \sim 3.5$. We note here that while a large enhancement in $\chi_p(T)$ at low T would imply a similar enhancement in $\gamma(T)$, such behavior would be difficult to discern from our $C(T)$ data due to the dominant phonon contribution.

In summary, detailed low-T transport and thermodynamic measurements imply that a highly correlated FL ground state develops beyond the superconducting dome in LSCO. The observation of an anomalously enhanced KW ratio suggests that *e*-*e* scattering in nonsuperconducting LSCO is much more intense than previously imagined and challenges existing claims that the ubiquitous T-linear resistivity observed in optimally doped cuprates is a signature of (strong) *e*-ph coupling.

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