The ground state of heavily-overdoped non-superconducting $La_{2-x}Sr_xCuO_4$

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We report detailed thermodynamic and transport measurements for non-superconducting $\text{La}_{1.7}\text{Sr}_{0.3}\text{CuO}_4$. Collectively, these data reveal that a highly-correlated Fermi-liquid ground state exists in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ beyond the superconducting dome, and confirm that charge transport in the cuprates is dominated at finite temperatures by intense electron-electron scattering.

The high- T_c cuprates (HTC) have emerged as one of the most formidable challenges facing the theory of correlated electrons in solids [1]. In particular, transport properties of the normal state present a number of uneasy paradoxes for the Fermi-liquid (FL) picture. Whilst it has been argued that conventional electron-phonon (eph) scattering might still 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 the density of carriers, that is, by moving towards the underdoped side. At sufficiently high carrier concentrations however, it has often been assumed that HTC 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 non-superconducting ground state in HTC. 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 $L_0 = 2.44 \times 10^{-8} \text{ W}\Omega/\text{K}^{-2}$. Surprisingly however, and at odds with a conventional FL picture, the WF relation was found to co-exist with a large linear resistivity term extending down to 0K. This dichotomy raises the question whether the field-induced 'normal state' in OD HTC, 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 Letter, we present in- (ρ_{ab}) and out-ofplane (ρ_c) resistivity, in- (κ_{ab}) and out-of-plane (κ_c) thermal conductivity, specific heat (C) and magnetic susceptibility (γ) 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}CuO₄ 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 50K, 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 reveals that intense e-e interactions persist beyond the superconducting dome and sheds important new light on the evolution of $\rho_{ab}(T)$ across the HTC phase diagram.

Seven bar-shaped samples (typical dimensions 3 mm x 0.6 mm x 0.6 mm) were prepared for either abplane (A1-4) or c-axis (C1-3) measurements from a large La_{1.7}Sr_{0.3}CuO₄ single crystal grown in an infra-red 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 95mK (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, whilst C(T) was measured between 0.6K and 10K 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 RuO₂ chips employed as one heater and two thermometers. $\rho(T)$ measurements were made on all seven samples using a conventional ac four-probe

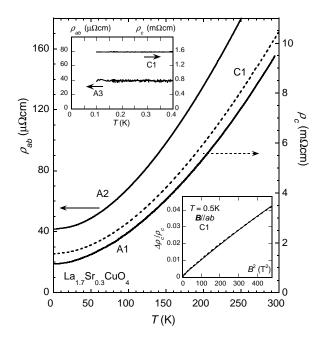


FIG. 1: Zero-field resistivity of A1, A2 (ρ_{ab} - solid lines) and C1 (ρ_c - dashed line). Upper inset: Low-T $\rho(T)$ of A3 and C1. Lower inset: $\Delta \rho_c/\rho_c$ versus B^2 for C1 at 0.5 K ($B\|ab$). The solid line is a fit to $\Delta \rho_c/\rho_c = 0.00010B^2$ - $3.1 \times 10^{-8}B^4$.

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

Fig. 1 shows $\rho(T)$ of A1, A2 and C1 below 300K. (All crystals reported here showed similar behavior). Note the similar metallic T-dependencies observed along both orthogonal directions, the different residual resistivity (ρ_0) values 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 ~ 65 at 300K to ~ 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 0.1A/cm^2 , well below the 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 = 0.5K. Inserting the fit to the low-field data (see caption) into the Boltzmann transport equation for a quasi-2D FL (see, e.g. [4]), we obtain an estimate of the in-plane electronic mean-free-path $\ell_{ab} \sim 145$ Å. Finally, using $k_F = 0.55 \text{Å}^{-1}$ for La_{1.7}Sr_{0.3}CuO₄ [5] and the "isotropic- ℓ " approximation [6], we obtain ρ_{ab0} = $21\mu\Omega$ cm, in good agreement with $\rho_0(A1)$.

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 non-

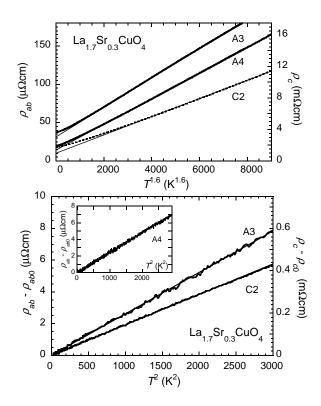


FIG. 2: a) $\rho(T)$ of A3, A4 (solid lines) and C2 (dashed line) versus $T^{1.6}$. Thin solid lines are provided as guides. b) Low-T $\rho(T)$ of A3 and C2 versus T^2 . Inset: ρ vs. T^2 for A4.

integer power-law resistivities were shown to extend from the lowest temperatures up to 1000K. Whilst $\rho(T)$ of our crystals follows very closely a $T^{1.6}$ dependence at elevated temperatures, the T-dependence clearly becomes stronger than $T^{1.6}$ as T drops below around 100K. Indeed, as shown in the bottom panel in Fig. 2, $(\rho \rho_0(T) = AT^2$ up to at least T = 55K for both current directions $((\rho - \rho_0)(T))$ for A4 is shown in an inset for clarity). Significantly, A has the same magnitude ($\sim 2.5 \pm 0.1 \ n\Omega \text{cm/K}^2$) 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, $(\rho - \rho_0)(T) = \alpha T + AT^2$ [3]) and implies that in La_{1.7}Sr_{0.3}CuO₄, charge transport can be understood in terms of an anisotropic 3D FL [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 $\kappa(\text{A1}) > \kappa(\text{A2})$, reflecting their respective ρ_0 values. The sizeable phonon term (κ_{ph}) in HTC has often been a major obstacle to the determination of κ_{el} , the electronic contribution to $\kappa(T)$ and the chief quantity of interest. Typically, κ_{ph} can only be unambiguously determined in the ballistic limit, below say 0.2K, where κ_{ph} is simply proportional to $\beta_3 T^3$, the low-T lat-

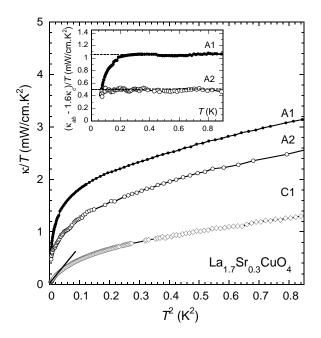


FIG. 3: Low-T thermal conductivity of A1, A2 and C1 plotted as κ/T vs. T^2 . Inset: $(\kappa_{ab} - 1.6\kappa_c)/T$ for A1 and A2. The dotted lines give κ_{el}/T as $T \to 0$.

tice heat capacity, as found here for C1 and indicated in the figure by a solid line. If κ_{ph} were isotropic, a more insightful approach might be to measure both κ_{ab} and κ_c on identical crystals and make use of the large σ_{ab}/σ_c anisotropy in HTC to delineate κ_{el} from κ_{ab} across the entire temperature range.

A quick inspection of Fig. 3 however reveals that κ_{ab}/T and κ_c/T are not simply offset from one another, implying either a T-dependent κ_{el}/T at low-T or anisotropy in $\kappa_{ph}(T)$ for in- and out-of-plane heat flow, due, for example, to anisotropy in the phonon velocities or e-ph coupling. The fact though that $\kappa/T(A1)$ and $\kappa/T(A2)$ are shifted relative to one another (except at the very lowest temperatures) suggests the latter, and simply by scaling $\kappa_c(T)$ by a constant factor (f = 1.6) and subtracting it from $\kappa_{ab}(T)$, we find, as shown in the inset to Fig. 3, that the resultant κ_{el}/T is indeed constant for both crystals and equal to 1.07 and 0.50 mW/cm.K² for A1 and A2 respectively (the dotted lines). This gives ρ_0 $=L_0T/\kappa_{el}=23$ (A1) and 49 (A2) $\mu\Omega$ cm, compared with the measured values of 19 and 42 $\mu\Omega$ cm. Given our somewhat simplistic way of extracting $\kappa_{ph}(T)$ from $\kappa_{ab}(T)$, it is encouraging to find that the WF law is satisfied for both crystals to within our experimental (geometrical) error. It should be stressed here that $\sigma_{ab0}(LSCO)$ is almost an order of magnitude smaller than in OD Tl2201 [3], thus making determination of κ_{el}/T more difficult.

The surprising feature of Fig. 3 is of course the strong downturn in $\kappa(A1)$ as $T \to 0$. (A slight downturn is also observed for A2 below 0.1K, though nowhere near as dra-

matic.) While slight deviations from the WF law can be attributed to additional anisotropy in κ_{ph} not yet accounted for, there is no assumption on $\kappa_{ph}(T)$ which can erase the sudden vanishing of κ_{el} below 0.2K for A1. 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 electrondoped cuprate $Pr_{2-x}Ce_xCuO_4$ (PCCO) [9]. By suppressing superconductivity with a high magnetic field, Hill et al. found that κ_{el}/T , whilst violating the WF law for T > 0.3K (in the sense that it clearly exceeds $L_0/\rho(T)$), rapidly vanishes as T falls below 0.3K. These two anomalous observations appear to have a common origin. The sample-dependent aspect of the downturn, however, suggests that it may be extrinsic in origin and unrelated to any exotic electronic behavior, particularly in view of the other, more conventional results obtained here for La_{1.7}Sr_{0.3}CuO₄ above 0.2K. 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}CuO₄, 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}CuO₄ to other strongly correlated systems. Electronic correlations in a FL are known to lead to an enhancement in the quasi-particle 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 $(A/\gamma_0^2 = a_0 = 10^{-5}\mu\Omega\text{cm}/(\text{mJ/Kmol})^2$ [11, 12]) and the Wilson ratio $(R_w = (\frac{\pi k_B^2}{3\mu_B^2}) \chi_p/\gamma_0$, 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 K < T < 10 K) to the expression $C = \alpha T^{-2} + \gamma_0 T + \beta_3 T^3 + \beta_5 T^5$ with $\alpha = 76~\mu\text{J.K/mol.}$, $\gamma_0 = 6.9~\text{mJ/mol.K}^2$, $\beta_3 = 0.1~\text{mJ/mol.K}^4$ and $\beta_5 = 0.7~\mu\text{J/mol.K}^6$. 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 to nuclear hyperfine or quadrupole splitting. The phononic $\beta_3 T^3$ term is comparatively small and gives rise to an elevated $\Theta_D = 550 \text{K}$, though β_3 may be made artificially low by a T-dependent electronic term [14].

The magnitude of γ_0 , though, is very robust in our measurements, 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 non-superconducting LSCO well off the so-called 'universal' line for strongly correlated metals indicated in Fig. 4. A similarly enhanced KW ratio, i.e. $A/\gamma_0^2 \geq 5a_0$, can also be inferred for OD Tl2201

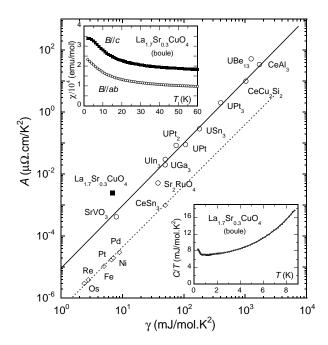


FIG. 4: Kadowaki-Woods plot of A, the coefficient of the T^2 resistivity, versus γ_0 , the electronic specific heat coefficient, for a variety of strongly correlated metals (adapted from [12, 13, 15]). Upper inset: Magnetic susceptibility $\chi(T)$ of large boule at 5T with B||c (solid squares) and B||ab (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.

 $(A=5.4 \text{ n}\Omega\text{cm}\text{K}^{-2} \text{ [3]}, \gamma_0 \sim 10 \text{ mJ/mol.K}^2 \text{ [17]})$, and indirectly for OD PCCO, $(A\sim 4 \text{ n}\Omega\text{cm}\text{K}^{-2} \text{ [18]}, \gamma_0 \sim 6.7 \text{ mJ/mol.K}^2 \text{ [19]})$. This observation reveals a new aspect of the physics of the cuprates and is the central result of this Letter. Whilst various explanations for the magnitude of the KW ratio in heavy fermion systems have been proposed, including a strong frequency dependence of the quasi-particle self-energy [12] and proximity to an antiferromagnetic instability [20], none have so far predicted deviations from a_0 as large as those found in the HTC. It is noted, however, that the effect of a strong momentum dependence in the scattering amplitude, thought to be a key feature of HTC [4], has yet to be taken into account [21] and efforts to include such an effect could prove highly illuminating.

As one moves across the HTC phase diagram towards the Mott insulator at half-filling, one expects e-e scattering to become even more intense. Thus, the gradual evolution from quadratic to linear resistivity in HTC must somehow 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}CuO₄ is ≈ 2 -3 times larger than that of La_{1.7}Sr_{0.3}CuO₄ at 300K, even though the Luttinger sum rule dictates that their carrier densities differ by

only a few percent. Moreover, given the relatively minor changes observed in the phonon spectra as a function of doping, one does not expect the strength of the e-ph interaction to change forcibly across the phase diagram. Thus, since e-ph scattering appears to give a negligible contribution to $\rho_{ab}(T)$ in La_{1.7}Sr_{0.3}CuO₄, it seems highly unlikely now that e-ph scattering can in any way account for the linear resistivity that appears (in a narrow composition range) around optimal doping. In the light of these results, we conclude that chronic (Umklapp) e-e scattering processes must dominate the normal state transport behavior of the cuprates across the entire accessible doping range. Such a scenario is consistent with the large increase in the quasi-particle lifetime below T_c observed in thermal and electrical conductivity measurements [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 χ_p [25] and an isotropic Curie-Weiss term (with $\Theta_C \sim 7.5 \mathrm{K}$), we obtain an average $\chi_p \sim 1.75 \times 10^{-4}$ 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 whilst a large enhancement in $\chi(T)$ 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 in La_{1.7}Sr_{0.3}CuO₄ have revealed for the first time that LSCO develops a highly correlated FL ground state beyond the superconducting dome. The observed enhancement in the KW ratio suggests that e scattering in non-superconducting LSCO is much more intense than previously imagined and raises serious question marks over previous claims that the ubiquitous linear resistivity observed in optimally-doped cuprates is a signature of (strong) e-ph coupling.

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