Anomalous normal state magnetotransport in an electron-doped cuprate

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We report magnetoresistance and Hall angle measurements of the electron-doped cuprate La$_{2-x}$Ce$_x$CuO$_4$ from 50 K to room temperature, expanding the transport phenomenology of this unconventional "strange metallic" system. We find a sharp transition in the field dependence of the magnetoresistance centered at 90 K in all dopings within the superconducting dome which may be associated with the onset of the high-temperature strange metallic phase. In addition, we find a non-Fermi liquid $T^4$ dependence of the cotangent of the Hall angle, further supporting the view that the high-temperature strange metallic phase lies outside the realm of Fermi liquid theory.

The normal state of the cuprate high-temperature superconductors has captivated the interest of the condensed matter physics community for the past decade while defying every attempt at theoretical explanation. The transport phenomena observed in these materials is believed to depart from the conventional Landau Fermi liquid theory of metals [1] and such "non-Fermi liquid" behaviors appear to be a common feature of high-temperature superconductors [2]. Consequently, it is reasonable to imagine that the mechanism of high-temperature superconductivity may naturally emerge from this non-Fermi liquid "strange metal" just as phonon-mediated superconductivity naturally emerges from a conventional Fermi liquid, making an understanding of this strange metallic state a potential stepping stone toward identifying the origin of high-temperature superconductivity.

The typical hallmark of the strange metallic state is the infamous linear-in-$T$ resistivity of the hole-doped cuprates, which persists over an anomalously large temperature range, from $T_c$ to 1000 K in some systems [3, 4]. However, the electron-doped compounds also exhibit a plethora of strange metallic behavior which differ sharply from the conventional properties of a Fermi liquid [5]. Further, these materials display two distinct regimes of strange metallicity with different behaviors at high and low temperatures, and which may or may not be of a common origin.

The high-temperature strange metallic phase of the electron-doped cuprates is characterized by a universal quadratic-in-$T$ resistivity, seen in all compounds and for all dopings [6, 7] from roughly 100 K to above 600 K [8]. This $T^2$ behavior is what one might naively expect for a Fermi liquid, although more thoughtful consideration of the large magnitude of the resistivity and the high temperature scale at which it is observed lead one to conclude that is in fact an extremely strange transport behavior [7], and is arguably stranger than the high-temperature linear-in-$T$ resistivity of the hole-doped materials, which could potentially be explained by electron-phonon scattering in a low carrier density system [9] and is a generic feature of most conventional metals.

At low temperatures, the nature of the strange metallic ground state is strongly doping-dependent and non-universal. Generically, the phase diagram of the electron-doped cuprates is dominated by a Fermi surface reconstruction (FSR) which occurs inside the superconducting dome [10–14] and is believed to be driven by short-ranged antiferromagnetic fluctuations [15]. For the material of interest in this study, La$_{2-x}$Ce$_x$CuO$_4$ (LCCO), this FSR occurs at cerium concentration $x = .14$ [16] (for reference, the SC dome extends from $x = .07$ to $x = .175$). In samples doped below the FSR (i.e. $x < .14$ for LCCO), the low-temperature resistivity exhibits an upturn, increasing with decreasing temperature [16, 17]. The origin of this upturn, also seen in hole-doped cuprates [18, 19], is not well understood, but is thought to be associated with the underdoped materials’ proximity to an antiferromagnetic insulating phase [20, 21].

The low temperature transport behavior of samples doped beyond the FSR ($x > .14$ in LCCO) has proven to be particularly intriguing. Remarkably, the resistivity in this region of the phase diagram varies linearly with temperature from a doping-dependent crossover temperature of the order of tens of Kelvin down to the lowest measured temperature of 30 mK when superconductivity is suppressed with an external magnetic field [22]. This is in stark contrast to the Fermi liquid expectation of a low-temperature $T^2$ resistivity, and is perhaps the most compelling evidence available for a non-Fermi-liquid ground state. In addition, it has recently been found that the low-temperature magnetoresistance (MR) of these overdoped samples is linear in magnetic field, in contrast to the conventional $H^2$ dependence expected for weak fields [23]. Further, the resistivity as a function of temperature and magnetic field obeys a scale-invariant functional form which, together with an anomalous logarithmic temperature dependence of the thermopower [24], is suggestive of quantum criticality [5, 23, 25].

Given the existence of these two regimes of strange metallicity the universal high-temperature $\rho \sim T^2$ and the low-temperature $\rho \sim T$ for dopings above the FSR it is reasonable to wonder whether they are continuously connected or are two distinct phases. The purpose of this work is to consider the intermediate temperature range at which a crossover between these two regimes may occur. Specifically, since all dopings of LCCO attain a $T^2$ resistivity by 70 K [7], we will be primarily interested in...
FIG. 1. (a) Magnetoresistance (MR) for an $x = .15$ sample measured up to 9 T. The crossing of the curves taken at 80, 90, and 100 K suggest the MR at 80 K grows more rapidly with field than at 100 K. (b) Kohler’s plot for same $x = .15$ sample. The curves for all temperatures studied below 80 K collapse onto one line, indicating that Kohler’s rule is obeyed. At 90 K Kohler’s rule is suddenly violated, and curves for 100 K and above again collapse onto one another. The bending of the high-temperature curves indicates a sub-quadratic dependence on magnetic field. Inset: Plot of the magnetic field exponent, $n$, of the MR as a function of temperature, obtained by fitting $\rho(H) = \rho_{H=0} + A H^n$. The exponent sharply changes from $n = 2$ to $n \approx 1.5$ at 90 K, where Kohler’s rule is violated. (c) Kohler’s plot for MR measurements taken between 80 and 100 K, where the exponent is rapidly changing. No curves collapse onto each other, and thus Kohler’s rule is violated everywhere in this temperature range. Inset: zero-field resistivity as a function of temperature over the same temperature range. Since this curve is continuous and essentially featureless, the change in the MR power law must be an intrinsically field-dependent effect.

the temperature range between 60 and 100 K. As shown in numerous prior works (see also the inset of Fig. 1c), the resistivity as a function of temperature is continuous throughout this temperature range, and we must thus consider other transport measurements to study a potential crossover. As evidenced by MR measurements, we report a universal (doping independent) transition occurring at around 90 K, and a puzzling but decidedly non-Fermi-liquid temperature dependence of the cotangent of the Hall angle.

All measurements are performed on $c$-axis oriented epitaxial thin films of LCCO grown via pulsed laser deposition on SrTiO$_3$ substrates. Details of the sample preparation can be found in the literature [16].

In Fig. 1a, we show representative data of the MR, $\Delta\rho/\rho_{H=0} \equiv [\rho(H) - \rho(0)]/\rho(0)$, for an $x = .15$ LCCO sample from 50 to 130 K. In particular, note that the curves taken at 80, 90 and 100 K cross one another, indicating the MR increases more rapidly with field at 80 K than at 100 K. Further, by inspection the MR at lower temperatures appears to be quadratic in field, while the higher temperature curves seem to have a slower field dependence. A useful lens through which to consider the MR behavior of a metallic system is Kohler’s rule, the statement that the MR should depend on the ratio of the mean free path to the cyclotron radius in a simple semiclassical picture. More formally, as can be seen from the Boltzmann equation, it is the statement that the MR depends only on the product of the magnetic field and scattering time, or more practically (assuming $\rho_{H=0} \sim \tau^{-1}$) is a function of only the ratio of the magnetic field to the zero-field resistivity [26],

$$\Delta\rho/\rho_{H=0} = F(H\tau) \approx F\left(\frac{H}{\rho_{H=0}}\right)$$

In Fig. 1b we assess the validity of Kohler’s rule in this system by plotting the MR against $(\mu_0 H/\rho_{H=0})^2$. For a conventional metal, one would expect the MR curves measured at each temperature to be linear on this plot and to all collapse onto one another. However, in LCCO one can see that such scaling holds below 80 K, is suddenly violated at 90 K, and then is satisfied again above 100 K, albeit with a sub-parabolic field dependence. Figure 1c shows MR curves taken at several temperatures between 80 and 100 K, wherein one can see that Kohler’s rule fails over the entirety of this temperature range.

To identify the origin of this abrupt violation of Kohler’s rule, we fit the field-dependent resistivity $\rho(H)$ to the form $\rho(H) = \rho_{H=0} + A H^n$. Plotting the field exponent $n$ as a function of temperature, as shown in the inset of Fig. 1b, one finds that $n$ sharply transitions from $n \approx 2$ (the conventional value of $n$ for a metal in a weak field) to $n \approx 1.5$ within a narrow temperature region centered at about 90 K.

Moving on from this single doping ($x = .15$) which we have used as an illustration of this phenomenon, we may consider other dopings within the SC dome. Similar Kohler scaling analyses show the same pattern for numerous dopings across the phase diagram: Kohler’s rule is obeyed below 80 K, violated over a limited temperature range, and obeyed with a slower field dependence above 100 K. This behavior is shown in Fig. 2 for highly underdoped and highly overdoped samples, as well as a
sample at the FSR doping. We note that due to the low-temperature resistivity upturn exhibited by underdoped samples (see the inset in the first panel of Fig. 2), Kohler’s rule is trivially violated at low temperatures in this doping range. Nonetheless, Kohler scaling is satisfied over a narrow temperature range, and a jump in this scaling slightly above 90 K is still seen as in the other dopings.

Measuring the MR and extracting the exponent \( n \) of the field-dependent resistivity for each doping, we find the same sharp transition at 90 K for all dopings within the SC dome, as shown in Fig. 3. Note that below the temperature range of interest in this work, samples doped below the FSR (\( x < .14 \)) have a negative magnetoresistance [20, 27] and samples above the FSR (\( x > .14 \)) display the aforementioned linear-in-\( H \) behavior [23]. But, for all dopings, the MR power law crosses over to a quadratic dependence by 50 K where our measurements begin. Samples outside the SC dome (\( x > .175 \)) were not considered in this study, owing to the presence of negative magnetoresistance, associated with the recently discovered ferromagnetic order [28] in this region of the phase diagram, which persists up to \( \sim 90 \) K, obscuring the analysis.

There are several puzzling features of this change in the MR power law. First, it occurs at a rather large temperature-scale: over three times the optimal superconducting critical temperature in this system. Secondly, it is quite sharp despite the high temperature at which it occurs, with the half-width of the feature being less than 10 K. The MR power law above the transition also takes an unconventional value of \( n \approx 1.5 \) which is not predicted by any standard theory of metals. Finally, the transition appears to be a universal feature of the LCCO phase diagram, occurring at roughly the same temperature for all dopings within the SC dome. Taking all of these observations together, it is reasonable to associate this feature with the only other universal property of the electron-doped cuprates, namely the strange metallic \( T^2 \) resistivity. Given that all dopings of LCCO have attained a \( T^2 \) power law of the resistivity by 90 K [7], at which the transition in the MR occurs, we speculate that this tran-
sition may in fact be a signature of the system’s entrance into the high-temperature strange metallic state.

In passing, we may also compare the observed behavior of the MR in LCCO to that of hole-doped cuprates in comparable temperature ranges. Prior studies of underdoped Hg1201 and LSCO [29], as well as overdoped Th2201 [30] find that Kohler’s rule (or some slightly modified version of it) holds below room temperature with an MR power law of \( n = 2 \), as one would expect for a conventional metal. Modulo the transition at 90 K, the situation in LCCO is somewhat similar, in that Kohler’s rule is satisfied for all temperatures except those at which the exponent is rapidly changing (see Fig. 1c). However, the presence of the transition and the unconventional \( n \approx 1.5 \) behavior of the MR above it differentiate the magnetotransport phenomenology of the electron-doped compounds from their hole-doped counterparts, and may be manifestations of a complex interplay between multiple strange metallic states presumably absent in the hole-doped materials, given that the universal slope of the linear-in-\( T \) resistivity suggests a single strange metallic phase in those compounds.

To supplement these MR measurements, in Fig. 4a we present measurements of the of the Hall coefficient for several dopings throughout the SC dome. Note that even at room temperature, the Hall coefficient has a nontrivial temperature dependence, departing from single-carrier Fermi liquid expectations. Moreover, \( R_H \) changes sign at high temperatures for dopings near the FSR, which naively suggests that both electron- and hole-like carriers may be relevant to the high-temperature transport properties of LCCO within this doping range. However, given the longstanding confusion over the meaning of the Hall coefficient in the cuprates, such a conclusion may very well be premature.

To further our characterization of the high-temperature strange metallic phase, in Fig. 4b the tangent of the Hall angle, \( \tan \theta_H \equiv \rho_{xy}/\rho_{xx} \) is shown for several dopings up to room temperature. Although the cotangent of the Hall angle, \( \cot \theta_H = \rho_{xx}/\rho_{xy} \) is the typical quantity of theoretical interest, the zeroes of \( \rho_{xy} \) which, as mentioned above, are present in some dopings up to 100-200 K, prevent the evaluation of this ratio. We note that \( \tan \theta_H \) is strongly doping dependent at intermediate temperatures, but by 200 K the curves for all dopings begin to collapse, which is again suggestive of a universal behavior of the high-temperature phase.

For the optimally doped \( x = .11 \) sample, \( \rho_{xy} \) does not change sign above 100 K, allowing for \( \cot \theta_H \) to be analyzed. As shown in the inset of Fig. 4b, \( \cot \theta_H \) exhibits a \( T^4 \) temperature dependence. In contrast, a Fermi liquid is expected to have \( \rho_{xx} \sim T^2 \) and \( \rho_{xy} \sim T^0 \), and thus \( \cot \theta_H \sim T^2 \), which is in fact one of the defining features of a Fermi liquid, and a behavior observed in the hole-doped cuprates [31, 32]. This clear \( T^4 \) dependence is thus a compelling indication that the high-temperature metallic phase of LCCO, and the electron-doped cuprates in general [33], is indeed a strongly correlated strange metal which cannot be understood within the context of Landau’s theory of Fermi liquids.

Altogether, we have demonstrated a sharp transition in the MR which occurs at 90 K, above which the MR varies as \( H^n \), with the anomalous exponent \( n \approx 1.5 \) for all dopings, and which we believe to be associated with the onset of the high-temperature strange metallic phase. We have also shown the non-Fermi liquid behavior of the Hall angle, \( \cot \theta_H \sim T^4 \), which when taken together with the well-established and extremely strange \( T^2 \) dependence of the high-temperature resistivity, unambiguously establishes this high-temperature regime to be a non-Fermi liquid phase.

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