Hidden strange metallic state in underdoped electron-doped cuprates

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The low-temperature linear-in-T resistivity of "strange metals," such as the metallic state of the cuprate high-temperature superconductors, has long been thought to be associated with a quantum critical point. However, recent transport studies of the cuprates have found this behavior persists over a finite range of overdoping. In this work, we report magnetoresistance and Hall effect results for electron-doped films of the cuprate superconductor $La_{2-x}Ce_xCuO_4$ (LCCO) for temperatures from 0.7 to 45 K and magnetic fields up to 65 T. For $x = 0.12$ and 0.13, just below the Fermi surface reconstruction (FSR) at $x = 0.14$, the normal state in-plane resistivity exhibits a well-known upturn at low temperature. Our new results show that this resistivity upturn is eliminated at high magnetic field and the resistivity becomes linear-in-temperature from ∼40 K down to 0.7 K. The magnitude of the linear coefficient scales with Tc and doping, as found previously [K. Jin, Nature(London) **476**, 73 (2011), T. Sarkar, Sci. Adv. **5**, eaav6753 (2019)] for dopings above the FSR. This striking observation suggests that the strange metal is not confined to a single "critical point" in the phase diagram, but rather is a robust universal feature of the metallic ground state of the cuprates.

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I. INTRODUCTION

The nature of the metallic normal state of the cuprate high-temperature superconductors is among the most actively discussed open problems in condensed matter physics. In particular, the anomalous linear temperature dependence of the resistivity observed at low temperatures [1,2] in these "strange metals" has defied theoretical description. This represents a fundamental departure from Landau's paradigmatic Fermi liquid theory, which is otherwise spectacularly successful at describing essentially all known conventional metallic states.

One well-established mechanism to realize a linear-in-temperature resistivity as $T\to 0$ is by tuning the system to a quantum critical point (QCP). The theory of transport near a 2D antiferromagnetic QCP is quite mature [3,4], predicting $\rho \sim T$ at the QCP, and beautifully describes a number of experimentally realized systems [5,6]. Such a picture is not to applicable to the cuprates, given that in many cuprate compounds the linear-in-T behavior is observed over an extended region of the phase diagram in LSCO [7], and in electron-doped (n-type) cuprates from the Fermi surface reconstruction doping up to the end of the superconducting dome [1,2]. Taken together, these observations seem to suggest that the origin of the linear-in-T resistivity in cuprates cannot be accounted for by an isolated quantum critical point, but rather are symptomatic of some exotic metallic ground state that exists over a range of dopings above some critical value.

The aim of the present work is to further characterize the nature and extent of this strange metallic phase in the electron-doped cuprate $La_{2-x}Cu_xCuO_4$ (LCCO) and $Pr_{2-x}Cu_xCuO_4$ (PCCO). The focal point of the electron-doped cuprate phase diagram occurs at a critical doping, $x_{FSR} = 0.14$ ($T_C = 19$ K) in LCCO, where the Fermi surface reconstructs from a large holelike Fermi surface for $x > x_{FSR}$ to a small Fermi surface with electron and holelike pockets for $x < x_{FSR}$. As mentioned above, a strange metallic $\rho{\sim}T$ behavior is observed down to 20 mK for all dopings above the Fermi surface reconstruction (FSR) to the end of the superconducting dome when superconductivity is suppressed with an applied magnetic field [1]. For dopings below the FSR, the low-temperature resistivity (in the normal state after supressing superconductivity with a c-axis magnetic field) exhibits an upturn [8,9]. The origin of this resistive upturn is not known but is widely believed to be due to prolific scattering off of antiferromagnetic spin [8–10]. This is supported by a theoretical model that suggests magnetic droplets can be formed by disorder in regions of the phase diagram that exhibit short range antiferromagnetic (AFM) order [10]. In this regime of the temperature-doping phase diagram, the normal state resistivity develops the well-known upturn. The resistivity tends to saturate as the temperature approaches zero showing that the normal state is a metal and not an insulator. Further, similar behavior is universally seen in underdoped hole-doped cuprates [11–18], i.e., the upturn is a generic feature of cuprate transport phenomenology.

The nature of this metallic state "hidden" beneath the resistive upturn in n-type cuprates was previously deemed to be a Fermi liquid (much like the metallic ground state of highly overdoped cuprates) based on an extrapolation of high-temperature transport data and extensive fittings [19]. In this work, we report direct measurements of the metallic ground state of underdoped electron-doped cuprates, and arrive at a very different conclusion. Namely, we find that the strange metallic phase, with its hallmark linear-in-T resistivity, persists even below the FSR. This is a new and

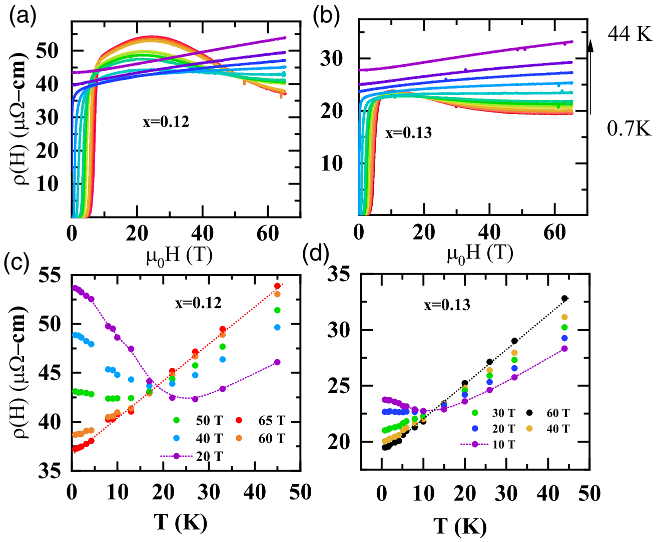

FIG. 1. Temperature dependent magnetoresistance ($\rho_{xx}(T,H)$) of LCCO for $x = 0.12$ (a) and 0.13 (b) ($H \perp ab$-plane). Black arrow indicates the increasing temperature direction from 0.7 to 44 K. (measured in 65 T pulsed field). In (c) and (d) the resistivity vs temperature is found from the (a) and (b) data, respectively. The H^*(the field where T_m vanishes) are $\sim$60 T for $x = 0.13$ and above 65 T for doping $x = 0.12$.

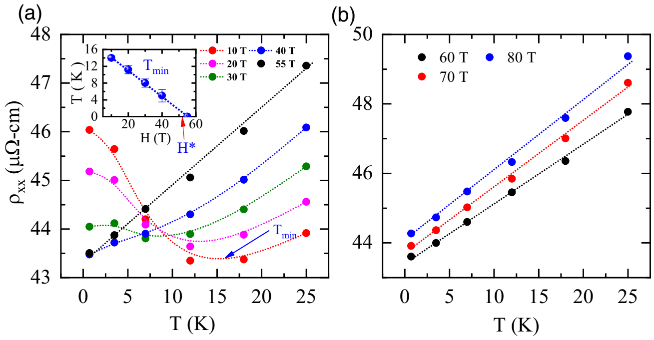

FIG. 2. (a) Transverse magnetoresistivity ($\rho_{xx}(T,H)$) vs temperature of $Pr_{2-x}Ce_xCuO_4$ for $x = 0.15$ (data taken from Ref. [22]). The dotted lines are guides to the eye. (Inset) The resistivity minima (T_m) vs field. The T_m is determined by taking the derivative of the dotted lines and H^* is the field where T_m vanishes. (b) Resistivity vs T at higher fields. The dotted lines are a linear fit with $\rho_{xx}(T,H) = \rho_{xx}(0,H) + A(x)T$, with $A(x) = 0.17$ (60 T), 0.19 (70 T), and $0.2(80\mathrm{T})\mu\Omega\,\mathrm{cm\,K^{-1}}$. Field is applied along the c axis for all data.

surprising discovery about the low-temperature normal state of the cuprates. We discuss the possible origins of this new experimental result later.

II. EXPERIMENTAL METHODS

The high field magnetic measurements were performed on $La_{2-x}Ce_xCuO_4$ (LCCO) films for two dopings ($x = 0.13$, 0.12) just below the Fermi surface reconstruction doping ($x = 0.14$). The films, of thickness about 150 nm, were grown using the pulsed laser deposition (PLD) technique on $SrTiO_3$ [100] substrates (5 × 5mm2) at a temperature of 750°C utilizing a KrF excimer laser. The films were post-annealed at 600° C in an oxygen partial pressure of about 1×10^{-5} torr for 30 minutes to remove the apical oxygen and induce superconductivity. The full width at half maximum of the peak in $d\rho_{xx}/dT$ of the films is within the range of 0.5 K, demonstrating the high quality of the samples. The LCCO targets have been prepared by the solid state reaction method using 99.99% pure La_2O_3, CeO_2, and CuO powders. The Bruker x-ray diffraction (XRD) of the films shows the c-axis-oriented epitaxial LCCO tetragonal phase. The thickness of the films has been determined by using cross-sectional scanning electron microscopy (SEM). The high-field 65 T measurement was performed by standard four-probe ac lock-in method at the National High Magnetic Field Laboratory (NHMFL) Pulsed Field Facility, Los Alamos National Laboratory and the 35-T dc field measurements were performed at the National High Magnetic Field Laboratory, Tallahassee DC field lab, Florida.

III. RESULTS

Figures 1(a) and 1(b) show the measured in-plane magnetoresistivity, $\rho_{xx}(T,H)$, of x = 0.12 ($T_C = 24\,\mathrm{K}$) and 0.13 ($T_C = 21\mathrm{K}$) LCCO samples up to fields of 65 T for numerous temperatures between 0.7 K and 44 K. The temperature dependence of the magnetoresistivity, extracted by taking cuts of each curve in Figs. 1(a) and 1(b) at a fixed field, is plotted in Figs. 1(c) and 1(d) for several different values of the field. At low fields, the low-temperature normal state resistivity develops the well-known upturn [8,9,20,21] mentioned above. The temperature at which the resistivity reaches a minimum, T_m, decreases as the field is increased, and eventually vanishes at a field which we will label H^*. At this field, the resistivity is linear in temperature, as one can see from the 65 T (red) curve in Fig. 1(a) and the 60 T (black) curve in Fig. 1(b).

The temperature dependence of the resistivity above H^*, in particular whether it remains linear or crosses over to another power law, cannot be ascertained for LCCO from our current data since $H^* \approx 65$ T was the maximum field available for the measurement. However, by re-analyzing our previous measurements of another electron-doped cuprate, $Pr_{2-x}Ce_xCuO_4$ (PCCO), we can address this issue. To compare with our results on LCCO, it is important to realize that the FSR occurs at $x = 0.17$ in PCCO (and NCCO). Figure 2 shows the temperature dependence of the magnetoresistance of an x = 0.15 PCCO sample for fields up to 80 T, taken from Ref. [22]. For this sample, $H^* \approx 55$ T and as seen in Fig. 2(b) the resistivity remains linear for all measured fields above H^*. Fitting these resistivity curves to $\rho_{xx}(T,H) = \rho_{xx}(0,H) + A(x)T$, we find that the coefficient of the T-linear resistivity $A(x)$ increases slightly with field from $0.17\mu\Omega\,\mathrm{cm\,K^{-1}}$ at 60 T to $0.2\mu\Omega\,\mathrm{cm\,K^{-1}}$ at 80 T. This slight increase may be a consequence of the emergence of a quadratic-in-field contribution to the magnetoresistance at temperatures above T_c that causes the magnitude of the magnetoresistivity to be larger at higher temperatures (see Fig. 3 of Ref. [2]).

In Fig. 3(a), we fit the resistivity vs. temperature curves for the $x = 0.12$ LCCO sample at 65 T and the $x = 0.13$ LCCO sample at 60 T to the form $\rho_{xx}(T,H) = \rho_{xx}(0,H) + A(x)T$, where $\rho_{xx}(0,H)$ is the resistivity at zero temperature and the

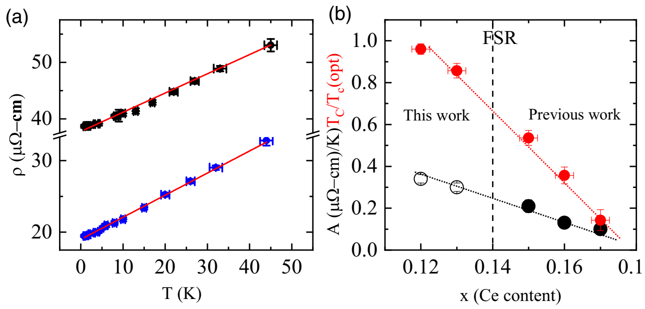

FIG. 3. (a) Resistivity vs temperature for LCCO x = 0.12 at 65 T (black data points) and x = 0.13 at 60 T (blue data points) from 700 mK to 45 K. The red solid line is a fit to $\rho_{xx}(T,H) = \rho_{xx}(0,H) + A(x)T$ (b) Previous work is reported in Refs. [1,2,4]. Open circles [from (a)] and solid black circles (taken from Ref. [2]) are the slopes, $(A(x))$, of the linear-in-T resistivity. The red circles are $T_c(x)$ normalized to the T_c at the optimal doping (26 K). Dotted lines are guide to the eye.

appropriate field. The slight low-temperature deviation from linearity in the $x = 0.12$ doping is likely a sign that 65 T is less than H^*, i.e., not quite sufficient to completely suppress the resistivity upturn.

The fitted slopes of the T-linear resistivity for the LCCO $x = 0.13$ and 0.12 are plotted as a function of doping in Fig. 3(b), along with previously measured values of A(x) for overdoped LCCO samples taken from Ref. [2]. Past work on overdoped LCCO has established that *A(x)* ∼ *1/x* and scales with the critical temperature for dopings above the FSR in the strange metal regime. Here, we find that *A(x)* for the underdoped $x = 0.13$ and 0.12 samples fall on the same *A(x)* ∼ *1/x* curve as the overdoped samples and scales the same way with T_c. Thus the linear-in-T resistivity reported here for underdoped samples appears to be of the same origin as that seen in overdoped samples.

In addition to our magnetoresistivity measurements, we report the high-field Hall coefficient as a function of temperature and magnetic field for LCCO, $x = 0.12$ and 0.13. It is known from our prior work on underdoped samples [9] that the low-field (below 14 T) Hall coefficient is peaked at a doping-independent temperature of order 10 K. Figure 4 shows the Hall coefficient measured from 2 to 80 K as a function of magnetic field up to 35 T. As shown in Fig. 4(b), as the magnetic field increases the low temperature peak in the Hall coefficient decreases and vanishes at high field, similar to how the resistivity minima vanishes with higher field (see Fig. 1). In Figs. 4(a) and 4(c), we see that above ∼10 K the magnitude of the Hall coefficient increases with increasing field and tends to saturate at high field. This indicates that the resistivity minima and Hall coefficient peaks are interlinked, as was suggested in the previous reports [9].

IV. DISCUSSION

To access the metallic ground state of the underdoped cuprates, we suppress the low-temperature resistive upturn with a large magnetic field, just as one routinely suppresses superconductivity in these materials. Naively, we imagine that the strong out-of-plane field repress antiferromagnetic spin, the scattering off of which is believed to be responsible for the upturn. Of course, given that the origin of the upturn is unknown, this interpretation is necessarily heuristic and further theoretical work will be needed to fully understand the microscopic mechanism responsible for the vanishing of the resistive upturn. That said, the energy scale of the field H at which the upturn disappears is qualitatively consistent with this simple physical picture, as will be argued below.

The temperature dependence of the high field resistivity and Hall coefficient (normal state) reported here for $x < x_{\mathrm{FSR}}$ LCCO is qualitatively similar to that of the $x > x_{\mathrm{FSR}}$ LCCO. Therefore it seems reasonable to speculate that the effect of strong magnetic fields is to move the location of the Fermi Surface Reconstruction (putative QCP) to lower Ce doping. This idea was proposed to explain some magnetic field effects in hole-doped cuprates [23,24]. However, our high field Hall coefficient at 2 K, $R_H \approx 0.5 \times 10^{-10}(\Omega - m)/T$ for the

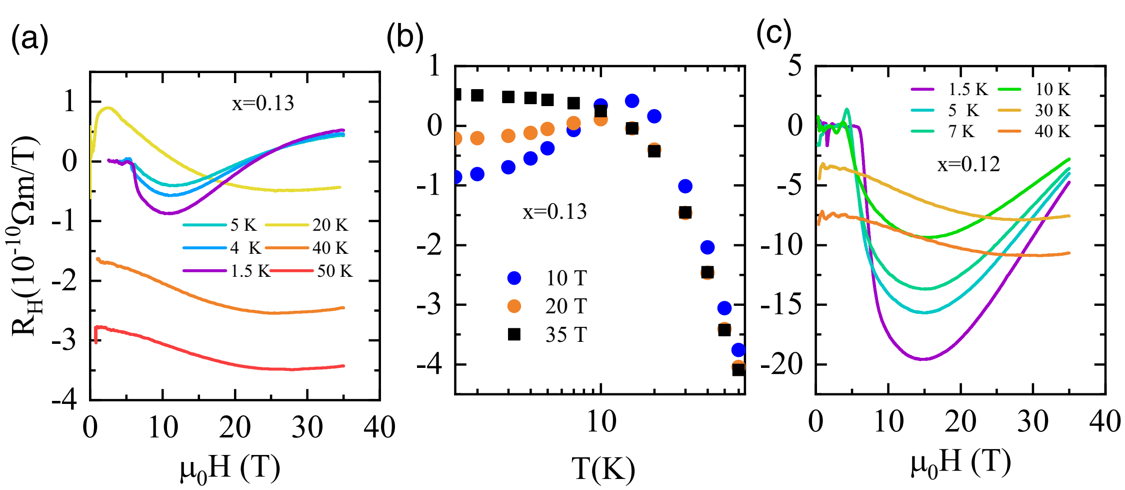

FIG. 4. (a) Hall coefficient vs field of LCCO for doping $x = 0.13$ at various temperatures. (b) The Hall coefficient vs T as a function of field [data taken from (a)]. (c) Hall coefficient vs field of LCCO for doping $x = 0.12$ at various temperatures. These data are found after subtracting any magnetoresistance component by measuring R_{xy} in magnetic fields from +35 to −35 T.

x = 0.13 doping [see Fig. 4(b)] is an order of magnitude lower than the large hole pocket R_H we found above the FSR [9]. Also, in this scenario, one would expect to find a large holelike FS at high fields. But, quantum oscillation experiments of electron-doped cuprates with $x < x_{\mathrm{FSR}}$ report low-frequency oscillations from the reconstructed small holelike pocket, even at 60 T [22,25,26], in conflict with such an interpretation. The high field quantum oscillation frequency also gives a small Fermi surface pocket size that is the same as that measured in zero-field by ARPES [27,28]. Thus it is unlikely that our findings can be thought of in terms of a shift in the position of the FSR.

Another possible explanation for our results is the magnetic field suppression of the in-plane, short range AFM, spin scattering that was proposed to be responsible for the resistivity upturn [7,9]. At a field of 50 T, the Zeeman energy ($g\mu_B B$) is approximately 60 K which is roughly $\sim 2\,T_{\mathrm{min}}$ (T_{min} is estimated by extrapolating the Fig. 2(a) inset plot to zero field, which is roughly 20 K). This means that the Zeeman energy $g\mu_B B$ at 50T is approximately the same as the energy corresponds to $2T_{\mathrm{min}}$ (at zero B). Thus an external 50 T field could greatly suppress the spin scattering responsible for the resistivity upturn. So, the field kills the antiferromagnetism and suppresses the resistive upturn but does not affect the position of FSR. This suggests the FSR may not be driven by short range antiferromagnetic order but driven instead by some other, possibly topological, order [27,28].

Next, we comment on the linear temperature dependence of the resistivity in LCCO and PCCO for $x < x_{\mathrm{FSR}}$ that emerges at high fields. This low-temperature linear-in-T behavior is the hallmark of the strange metal state observed in both electron- and hole-doped cuprates [21]. Such a state is observed in electron-doped cuprates for all dopings between the FSR doping and the end of the SC dome. After application of a large external field our results indicate that the underlying metallic ground state of $x < x_{\mathrm{FSR}}$ electron-doped cuprates is also a strange metal (at least for dopings near the FSR). Another feature of this strange metal state is a linear-in-H magnetoresistance at low temperatures, as was found for $x > x_{\mathrm{FSR}}$ LCCO films (2). Although our present experiments on LCCO x = 0.13 did not go to high enough field to measure this, we note that higher field experiments on a related n-doped cuprate did observe a linear-in-H magnetoresistance from 55 − 90 T for temperatures below 30 K [29]. Moreover, the fact that the coefficient of the linear-in-T resistivity scales with doping in the same manner as $x > x_{\mathrm{FSR}}$ samples (see Fig. 3) further suggests that the "hidden" strange metal ground state of $x < x_{FSR}$ samples is of the same origin as the strange metal state found on the $x > x_{\mathrm{FSR}}$ side of the phase diagram.

Our conclusions are in stark contrast to some prior work, which argued the normal ground state of electron-doped cuprates was best described as a Fermi liquid [19]. However, we note that this prior work was done at zero magnetic field and relied on an uncertain subtraction of an estimated upturn resistivity, whereas our work here is a direct measurement of the metallic ground state hidden underneath the resistivity upturn.

Our results indicate that a strange metallic ground state is present in the electron-doped cuprates for all dopings within the superconducting dome and is thus a universal feature of the electron-doped cuprates. Such a strange metal state is observed in hole doped LSCO [7], Bi2201 [30], and Tl2201 [31] for dopings above the pseudogap end point to the end of the SC dome. This is in stark contrast to many unconventional superconductors [5,6] where linear-in-T resistivity is observed only at a single, ostensibly quantum critical, doping. Consequently, this universality poses a challenge to many developing theories of strange metallic transport, in particular those which attribute the linear-in-T resistivity to quantum critical points. Further, our results demonstrate that strange metallic transport, whatever its origin, is largely insensitive to the Fermi surface character, in that it is observed on either side of the FSR where the Fermi surfaces vary significantly.

The Hall effect is another unexplained anomalous property of the cuprates. Many proposals have been made (for example see [30,32–35] and references therein) but there is no consensus yet. Here, we discuss the Hall coefficient in our two underdoped (i.e., below the FSR) LCCO films and show that the field and temperature dependence is inconsistent with conventional Boltzmann theory.

From the known fermiology [25–27] of underdoped electron-doped cuprates, consisting of a small holelike pocket and a large electronlike pocket, we may compare our Hall coefficient measurements to the standard two carrier Boltzmann transport model [36]. The components of the resistivity tensor are given by

$$\rho_{xy} = BR_H = \frac{1}{e}\frac{\left(n_h\mu_h^2 - n_e\,\mu_e^2\right) + \mu_h^2\mu_e^2 B^2(n_h - n_e)}{(n_h\mu_h + n_e\mu_e)^2 + \mu_h^2\,\mu_e^2\,B^2\,(n_h - n_e)^2}B \tag{1}$$

$$\rho_{xx}(B) = \frac{1}{e}\frac{(n_h\mu_h + n_e\,\mu_e) + \left(n_e\,\mu_e\mu_h^2 + n_h\,\mu_h\mu_e^2\right)B^2}{(n_h\mu_h + n_e\mu_e)^2 + \mu_h^2\,\mu_e^2\,B^2\,(n_h - n_e)^2} \tag{2}$$

where $n_h(\mu_h)$ and $n_e(\mu_e)$ are the carrier density (mobility) of electrons and holes, respectively.

However, the Hall coefficient does not fit with conventional two carrier Boltzmann. We find the fits to be inconsistent with conventional two carrier Boltzmann transport as argued below.

We compare the sign of R_H in the high field limit where Eq. (1) give a Hall coefficient, $R_H = \rho_{xy}/H = \frac{1}{e}\frac{1}{(n_h - n_e)}$. This equation suggests R_H should be negative since $n_e > n_h$. In contrast, we find that, at low temperatures (<10 K) and high field, R_H is positive (see Fig. 4(a) and Fig. 6 of Ref. [26]). In addition, the low-temperature high field Hall coefficient is strongly field dependent as oppose to conventional two carrier system where one would expect field independent Hall coefficient at high field. This suggests a possible Fermi surface instability due to the antiferromagnetic spin suppression at high field. A prior study on PCCO films also reported that the high field Hall coefficient does not fit with conventional two carrier Boltzmann transport for dopings just below the FSR [37].

The high field magnetoresistance (see the high field regime of Refs. [22,29]) is linear-in–H, which is also inconsistent with two-carrier transport where a quadratic field dependence magnetoresistance is expected in a conventional two-band model. Thus the normal state Hall coefficient and the normal state MR for LCCO $x = 0.12$, 0.13 doped films (just below the FSR at $x = 0.14$) exhibit an anomalous strange metal behavior. A modified model with consideration of field dependent spin scattering might explain our data; however, the development of such a theoretical model is outside the scope of the present experimental work.

V. SUMMARY

We have performed low-temperature, ab-plane resistivity and Hall-effect measurements of the electron-doped cuprate $La_{2-x}Ce_xCuO_4$ for dopings $x = 0.12$ and 0.13 and $Pr_{2-x}Ce_xCuO_4$ for doping $x = 0.15$ (both just below the Fermi surface reconstruction) at high magnetic fields. These strong fields suppress the low-temperature resistivity upturn to reveal a linear-in-T resistivity whose magnitude scales with that of the linear-in-T resistivity found at higher doping. This result implies that the normal metal state hidden beneath the resistivity upturn is the same strange metallic state observed in overdoped samples. The most accredited picture for cuprates is that linear-in-T resistivity at low temperature is only expected at a quantum critical point (QCP). But, our work presented here shows a strange metallic state for doping below the Fermi surface reconstruction (FSR). Along with prior studies of doping above the FSR (1, 21), this work strongly suggests that the low-temperature linear-in-T resistivity, the hallmark of the strange metal state, is a universal feature of the cuprates within the SC dome. This newfound ubiquity of the strange metallic state, and its apparent insensitivity to Fermi surface reconstruction, represents a significant development in our understanding of the cuprate phase diagram.

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