

Quantum critical scaling at the edge of Fermi liquid stability in a cuprate superconductor

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Edited by Douglas J. Scalapino, University of California, Santa Barbara, CA, and approved March 20, 2012 (received for review December 20, 2011)

In the high-temperature cuprate superconductors, the pervasiveness of anomalous electronic transport properties suggests that violation of conventional Fermi liquid behavior is closely tied to superconductivity. In other classes of unconventional superconductors, atypical transport is well correlated with proximity to a quantum critical point, but the relative importance of quantum criticality in the cuprates remains uncertain. Here, we identify quantum critical scaling in the electron-doped cuprate material $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ with a line of quantum critical points that surrounds the superconducting phase as a function of magnetic field and charge doping. This zero-temperature phase boundary, which delineates a metallic Fermi liquid regime from an extended non-Fermi liquid ground state, closely follows the upper critical field of the overdoped superconducting phase and gives rise to an expanse of distinct non-Fermi liquid behavior at finite temperatures. Together with signatures of two distinct flavors of quantum fluctuations, these facts suggest that quantum criticality plays a significant role in shaping the anomalous properties of the cuprate phase diagram.

A longstanding issue in the quest to understand high-temperature superconductivity in the cuprates is in regard to the nature of the underlying ground state. Exotic transport properties (1, 2) are widely considered to arise due to the nontrivial consequences of quantum criticality (3–6), resulting in a strongly correlated electronic ground state that underpins the infamous phase diagram of the cuprates (7). The recent observations of quantum oscillations in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (8) have provided a significant advance to our knowledge of the progression of this ground state through the cuprate phase diagram. The presence of small Fermi surface (FS) pockets distinct from the large FS structure observed in overdoped cuprates (9) requires the existence of an FS reconstruction, which logically occurs at a quantum phase transition between ground states that modify the symmetry of the Brillouin zone. With the origin of superconductivity still under hot debate, how the phase diagram is “shaped” by the evolution of these ground states remains a crucial question.

AN FS transformation has also been directly observed in the electron-doped cuprates as a function of doping, for instance as observed in $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (10, 11). This evolution is consistent with several indications of a quantum critical point associated with the suppression of antiferromagnetic order near optimal doping for superconductivity and the appearance of a Fermi liquid (FL) ground state on the overdoped side. With relatively low upper critical field values, the electron-doped cuprates allow for a unique opportunity to study the underlying ground state of the phase diagram in much detail (12). $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ (LCCO) is particularly unique in that its superconducting (SC) “dome” is centered at relatively lower Ce concentrations (13), making it possible to study the complete suppression of superconductivity by both doping and magnetic field. One of the most extraordinary characteristics of the cuprates is the hallmark temperature-linear resistivity, which was shown in LCCO to persist over three decades in temperature and to have a strong correlation with the pairing strength itself (14). Here, we study in detail the effects of applied magnetic field on LCCO, using the selective response of spin fluctuations and superconductivity to magnetic field and

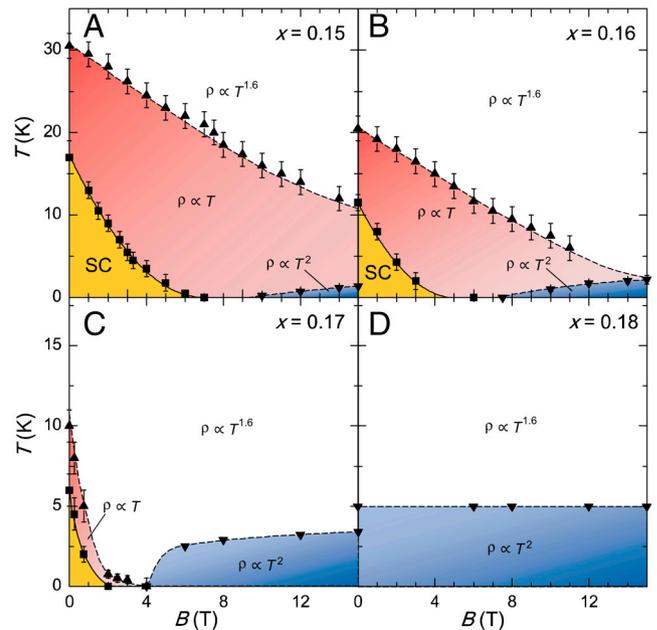


Fig. 1. Doping evolution of magnetic field-temperature phase diagrams of $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$. (A–D), The magnetic field dependence of the evolution of superconducting (yellow), Fermi liquid (blue), and non-Fermi liquid (red, white) ground states of the electron-doped cuprate system $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ is shown for several electron doping levels (x). These constant-doping magnetic field (B) temperature (T) phase diagrams illustrate the interplay of two distinct transport scattering rates, represented by both $\Delta\rho \propto T$ (red regions) and $\Delta\rho \propto T^{1.6}$ (white regions) power laws, that envelope the superconducting state and characterize the non-Fermi liquid behavior emanating from the quantum critical points marking the onset of the Fermi liquid state. The cross-overs between $\Delta\rho \propto T$ and $\Delta\rho \propto T^{1.6}$ scattering behavior dramatically shift with doping along with the onset field of the Fermi liquid state ($\Delta\rho \propto T^2$), indicating a strong dependence on both doping and magnetic field that persists with doping toward a dominant, field-independent state at $x = 0.18$ (D).

charge doping to segregate a complicated mixture of behaviors into two distinct signatures of criticality.

Results and Discussion

The nonsuperconducting FL ground state of overdoped LCCO can be readily accessed by either of two ways: doping in electrons beyond a critical value x_c , or increasing magnetic field above a critical value B_c that is greater than the superconducting upper

Author contributions: N.P.B., R.L.G., and J.P. designed research; N.P.B., K.J., K.K., and J.P. performed research; N.P.B., K.J., K.K., and J.P. analyzed data; and N.P.B., K.J., R.L.G., and J.P. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

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This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1120273109/-DCSupplemental.

Table 1. Definition and range of transport power law and critical exponents

Exponent	Determined by	Range of applicability	Value
n	power law fits to $\rho(T)$ data	temperature range depends on (x, B) (Fig. 1)	$1 < n < 2$
α	divergence of A_2 coefficients as B_c is approached from high B	all (x, B) exhibiting a Fermi liquid ground state	0.38 ± 0.01
β	divergence of A_2 coefficients as x_c is approached from high x	all (x, B) exhibiting a Fermi liquid ground state	0.72 ± 0.05
γ	collapse of $\rho(T)$ data for constant x at different values of B	all (x, B) exhibiting a Fermi liquid ground state, from 20 mK to upper limit of T^n behavior	$=\alpha(2-n), \beta(2-n)=0.4 \pm 0.1(x = 0.15)$ $=1.00 \pm 0.02(x = 0.17)$

The dynamical range of the fitted parameter over which each exponent has been determined is shown explicitly in Fig. 1 for the transport power law exponent n , where temperature ranges of best fit are dependent on doping and field values (e.g., for $x = 0.15$, $n = 1$ for three decades of T in the range $0.020 < T < 20$ K). For the critical and scaling exponents, the dynamical ranges are shown explicitly in Fig. 4 and listed here for critical exponents α ($0.01 < \Delta B/B_c < 3$) and β ($0.025 < \Delta x/x_c < 0.20$), as well as the scaling exponent γ ($0.06 < \Delta B'/T < 10$ for $\gamma = 0.4$, and $0.1 < \Delta B'/T < 10$ for $\gamma = 1$).

fluctuation (SF) region defined by the $n = 1.0$ scattering behavior that dominates a substantial range of temperature, magnetic field, and doping. Of course, the inception of superconductivity likely consumes much of the entropy associated with such a state (27), filling in most of the SF phase space as shown in Fig. 2. However, as shown in Fig. 2B, a tantalizing glimpse of a possible non-Fermi liquid phase (NFL) may be present between the SC upper critical field B_{c2} and B_c , where an extended range of $T = 0$ NFL behavior endures much like in other anomalous systems (28–30).

Thus, at $x = 0.15$, the $n = 1.0$ scattering mechanism is dominant, extending to the zero-temperature limit once B_{c2} is surpassed, and the resultant $\Delta B/T$ scaling obeys the expected self-consistency in a wide range of fields and temperatures reaching up to the SF scale T_1 . However, upon increasing doping from $x = 0.15$, the SF energy scale is dramatically reduced both in temperature and in field, with both scales terminating at the critical doping $x_c = 0.175$ where both T_1 and B_1 approach zero together with T_c and T_{FL} . Given the intimate correlation between T_1 and T_c in zero field (14), the discrepancy between their magnetic field dependence is all the more remarkable. It indicates that magnetic field does not destabilize superconductivity by destroying the mediating spin fluctuations, but rather through more mundane orbital effects. For instance, at $x = 0.15$, the upper temperature limit of the SF region, denoted as T_1 , is much more robust against magnetic field than T_c itself, extrapolating to a zero-temperature field scale B_1 that far surpasses B_{c2} (Fig. 2B). But at higher doping, T_1 and T_c are both suppressed at an almost equal rate toward zero close to B_c , and the $n = 1.6$ power law characterizes the dominant scattering rate at temperatures directly above the quantum critical point. For instance, in the special case of $x = 0.17$ at 4 T (Fig. 2C and *SI Text*), this power law persists to at least 13 K, spanning at least three decades in temperature when it is the dominant scattering mechanism.

This correspondence underscores two major points. First, the magnetic field-induced divergence, critical scaling and the NFL scattering temperature dependence can be understood within a self-consistent framework. Second, the fact that this self-consistency adjusts according to which scattering is dominant is evidence for critical behavior arising from two origins—two sets of anomalous scattering, two forms of scaling and self-consistent critical exponents. Clearly, there are two distinct scattering behaviors that respond differently to doping and magnetic field, and the competition of these two scattering mechanisms is directly borne out in the temperature dependence of resistivity throughout the field-doping phase diagram. With the $n = 1.0$ power law likely arising from scattering with fluctuations associated with the antiferromagnetism of the parent compound, the $n = 1.6$ power law appears to be a distinct signature of a second type of quantum critical fluctuation. Interestingly, this power law is strikingly simi-

lar to that observed in the hole-doped cuprates $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (1) and $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$ (31) in the vicinity of x_c , suggesting the quantum critical endpoint of the SC phase may give rise to fluctuations that cause this particular anomalous scattering behavior. In fact, recent measurements of both $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (32) and $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$ (33) indeed show quantum critical behavior originating from the end of the SC dome, pointing to a universal nature of the quantum phase transition separating the superconducting and Fermi liquid ground states. The possibility of calculating a nonperturbative critical theory of such fluctuations for a disorder-driven SC quantum critical point (34) shows promise for confirming such a scenario.

Clearly, quantum criticality plays a significant role in shaping the phase diagram of the electron-doped cuprates, both in optimizing the superconductivity as well as limiting its extent. The ensuing picture is that two proximal quantum critical points compete in the cuprate phase diagram. The first, positioned near optimal doping, gives rise to spin fluctuations that stabilize unconventional superconductivity. The second, at $B_c(x)$, owes its very existence to the first because it is born of the suppression of superconductivity and the emergence of the normal FL state. The result is a complex but tractable interplay of competing quantum critical fluctuations that conspire to shape the phase diagram that has become the ubiquitous signature of high-temperature superconductivity.

Methods

Samples. The c -axis-oriented LCCO films were deposited on (100) SrTiO_3 substrates by pulsed laser deposition utilizing a KrF excimer laser. The annealing process for each Ce concentration was optimized such that samples showed the narrowest SC transition widths or metallic behavior down to the lowest measured temperature (20 mK), whereas nonoptimized samples usually showed an upturn at low temperature, as previously reported. The films were patterned into Hall bar bridges using photolithography and ion milling techniques. Several samples of each concentration were studied to ensure that the data are representative.

Measurements. Electrical transport measurements at temperatures greater than 2 K were carried out in a commercial cryostat equipped with a 14 T magnet, whereas lower temperature measurements down to 20 mK were performed in a dilution refrigerator equipped with a 15 T magnet. Data from the two platforms were measured with overlapping temperature ranges. Current was applied in the ab plane while the magnetic field was applied along the c axis for all the measurements.

ACKNOWLEDGMENTS. We thank A. Chubukov, V. Galitski, J. Schmalian, L. Taillefer, and C. M. Varma for their discussions. This research was supported by the National Science Foundation under DMR-0952716 (to J.P. and K.K.) and DMR-1104256 (to R.L.G.) and the Maryland Center for Nanophysics and Advanced Materials (K.J. and N.P.B.).

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Supporting Information

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SI Text

Scaling of the Quadratic Coefficient A_2 of the Resistivity in the Fermi Liquid State. Fits to the electrical resistivity of the form $\rho = \rho_0 + A_2 T^2$ were performed on the high field side of B_c , i.e., in the field-induced Fermi liquid (FL) ground state. As a function of magnetic field, the data scale with identical critical exponent $\alpha = 0.38$. This analysis was performed on data from multiple samples at each concentration. In order to plot the data together in Fig. 4A, the absolute values of the coefficients for each sample were scaled by a constant value, which maintains the integrity of the scaling analysis. The necessity for rescaling is expected because of the sensitivity of the scattering to sample dependence beyond experimental control, which makes the success of the A_2 scaling all the more remarkable. A similar approach was used to put together Fig. 4B. Note that, experimentally, the step size is much coarser in the doping direction, and the uncertainty is larger due to the aforementioned sample dependence.

Resistivity Scaling Above the Fermi Liquid Boundary B_c . The scaling of $\rho(T)$ reflects the fact that the resistivity $\Delta\rho$ can be described generally as a function $A_2 T^2 f(\Delta B^\gamma/T)$, where $\Delta B = B - B_c$, that is applicable to scattering in both Fermi liquid ($\Delta\rho \propto T^2$) and non-Fermi liquid (NFL) ($\Delta\rho \propto T^n$) regions. In this framework, the T^n behavior in the NFL region stems from anomalous temperature dependence in A_2 , which is by definition a constant in temperature in the FL state. The resultant picture is that T_{FL} separates the FL state at high magnetic field and low temperature from the NFL region at low magnetic field and high temperature,

consistent with the magnetic field dependence of T_{FL} (Fig. 1C). This picture also suggests that upon crossing T_{FL} the dominant energy scale is transferred from temperature to magnetic field, which implicitly suggests that any dominant energy scale, such as Fermi energy, is absent.

The exponents α , γ , and n are related as $\alpha = \gamma(2 - n)$ by considering the following asymptotic limits: (i) Fermi liquid ($T \ll \Delta B$): $\Delta\rho = A_2(B)T^2$. In this limit, $\Delta\rho/A_2(B)T^2 = 1$ and thus $f(\Delta B^\gamma/T) \rightarrow 1$. (ii) Non-Fermi liquid ($T \gg \Delta B$): $\Delta\rho = A_n T^n = A_2(B)T^2 \times (\Delta B^\gamma/T)^{2-n}$. Our data show that when $n < 2$, $\Delta\rho < A_2(B)T^2$ and thus $f(\Delta B^\gamma/T) < 1$. Note that it is possible to define $A_2'(B, T) = A_2(B) \times (\Delta B^\gamma/T)^{2-n}$, or in other words, explicitly add a temperature dependence to A_2 , which is a constant in temperature in the Fermi liquid state. However, from Fig. 4A we already know that $A_2 \propto \Delta B^\alpha$, and because A_2 and A_2' must have the same magnetic field dependence, it follows that $\gamma(2 - n) = \alpha$.

For $x = 0.17$, scaling is satisfied using an exponent $\gamma = 1.0 \pm 0.02$, so $\alpha = 0.38$ forces $n \approx 1.6$. For $x = 0.15$, scaling is satisfied using an exponent $\gamma = 0.4 \pm 0.1$, so $\alpha = 0.38$ forces $n \approx 1.0$ (Fig. S4).

The plots in Fig. 4 show the difference between Fermi liquid and non-Fermi liquid behavior. In the Fermi liquid state, $\Delta\rho/A_2 T^2 = 1$ by definition, and the slope of the scaled curve is zero. In contrast, in the non-Fermi liquid regime the slope of the scaled curve is positive, reflecting the notion that A_2 is no longer a constant.

