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Interlayer magnetotransport in the overdoped cuprate $Tl_2Ba_2CuO_{6+x}$: Quantum critical point and its downslide in an applied magnetic field

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Fundamental explanations of high-temperature (high-T_c) superconductivity must account for the profound differences in the properties of the "normal" (nonsuperconducting) state at the two extremes of charge doping: heavy and light. On the light doping side, its properties clearly violate the standard Fermi-liquid theory of metals. The key to the nature of superconducting pairing lies in understanding the transition to a conventional behavior on the overdoped side. We report a convergence of the pseudogap energy scale and the boundary that separates unconventional from a conventional metal in the zero-temperature limit, both boundaries framing a V-shaped area of "strange metal" state in the temperature-doping phase space. By accessing the low-temperature regions of the phase diagram via a high-field interlayer magnetotransport in heavily doped $Tl_2Ba_2CuO_{6+x}$, we show that the pseudogap boundary has the hallmarks of a quantum phase transition with a zero entropy jump. The critical doping (linkage) point consistently downshifts with magnetic field in unison with the suppression of T_c, suggesting that quantum critical fluctuations that destabilize the pseudogap are connected to the superconductivity with high-T_c.

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I. INTRODUCTION

The link of the pseudogap state in the cuprates[1]—the anomalous partially gapped electronic excitations that persist to energy scales much higher than the superconducting transition temperature T_c—to the superconductivity with high T_c is still highly uncertain.[2,3] Even from a most direct angularly resolved photoemission spectroscopy (ARPES) data,[4,5] contradictory conclusions have been drawn. In theory, pseudogap has been considered either as a precursor to superconductivity[6,7]—a giant high-temperature phase fluctuation regime, with the suprconducting coherence established by chilling to the superconducting transition temperature T_c, or as a distinct competing phase[8–10] whose quantum-critical fluctuations could act to promote the superconducting state. The latter view requires that there be a zero-temperature (quantum) phase transition[11] as the superconductor is doped with charge carriers, either electrons or holes.

Demonstrating a transition in the $T \rightarrow 0$ limit is not trivial. It requires evidence that there is a critical point with three different states of matter around it: an ordered phase, a phase whose properties are dominated by quantum critical fluctuations, and a conventional Fermi-liquid (FL) metal phase. To track these "normal" phases, a full and controlled doping range, preferably in a cuprate with structure and homogeneity unchanged, ought to be explored. One major difficulty with this comes from the material constraints.[12,13] Another, is that at low temperatures, superconductivity—which itself arguably cannot be understood without understanding the "metallic" states—intervenes. Indeed, while several recent observations report symmetry breaking[14–17] in the pseudogap phase, the vanishing of the pseudogap at a quantum critical point (QCP) is suggested only by extrapolation from fairly high temperatures.

Here, using a confluence of material properties and their dependence on doping and magnetic field, we access the phase diagram in the low-temperature limit. We have found that the structurally simple $Tl_2Ba_2CuO_{6+x}$ (Tl-2201) having a single CuO_2 layer per unit cell, is ideal in this regard.[18] It is clean [optimal T_c is above 90 K, as compared, for example, to $\lesssim 30$ K in a relatively disordered $La_{2-x}Sr_xCuO_4$ (LSCO) (Ref. 19)] and can be heavily overdoped (see Fig. 1) by changing oxygen content.[20] With Tl-2201—by exposing extended regions of normal state as superconductivity is suppressed with high (up to $H \sim 65$ T) magnetic fields—we can follow the phase diagram to nearly the end of the superconducting dome. We have previously discovered that the non-Fermi-liquid (n-FL) normal-state transforms into a conventional metal at high magnetic fields,[21] but this left unresolved the critical issue of the connection of the superconductivity to the pseudogap, whose peculiar sensitivity to doping is quite unlike the parabolic doping dependence of T_c.

The principal result of this study is demonstrating that the crossover boundary to the Fermi liquid, T_{FL}, and the onset temperature T^* of the pseudogap, merge at a critical doping composition in the limit $T \rightarrow 0$, identifying a quantum critical doping point p_c. In magnetic field, as the superconducting transition temperature T_c is reduced, p_c shifts in magnetic field to lower doping, tracking the suppression of T_c in the temperature-doping (T-p) phase space. The intimate low-temperature linkage of the $T^*(p)$ and $T_{FL}(p)$ boundaries and the field-linear shift of $p_c(H)$ pinned to the T_c dome, together with the scaling and diverging features, consistently imply

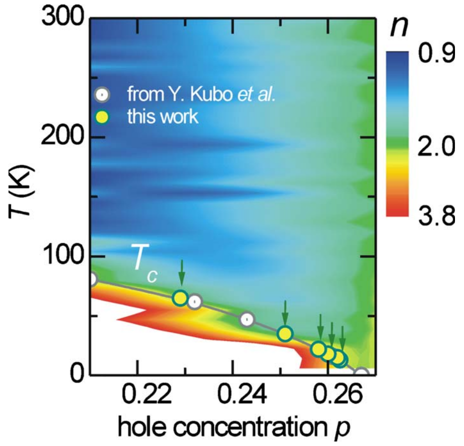

FIG. 1. (Color online) Experimental phase diagram of the cuprates vs the number of doped holes per Cu ion from charge transport. The in-plane resistivity $\rho_{ab} \propto T^n$ in Tl-2201 shows a smooth evolution with doping from the non-Fermi-liquidlike power law with $n \sim 1$ to a very standard T-squared dependence where $T_c(p)$ becomes zero (Ref. 20). T_c's of the crystals used in this work are indicated by the arrows.

that the singular regime between the pseudogap and the Fermi liquid is the critical fluctuation regime tied to superconductivity with high T_c.

II. EXPERIMENTAL

Single crystals of $Tl_2Ba_2CuO_{6+x}$ were grown by a flux method.[18] We used a series of homogeneous overdoped crystals with transition temperatures T_c spanning a wide range from 15 to 65 K. The hole concentration level p was determined using a well-established phenomenological relation[13,20] between T_c and p, $T_c/T_c^{\max} = 1 - 82.6(p-0.16)^2$.

The c-axis resistivity $\rho_c(T,H)$ was measured either in dc fields using the 45 T hybrid magnet at NHMFL, Florida,[21] or in a pulsed magnet at the National High Magnetic Field Laboratory (NHMFL in Los Alamos), where resistivity was recorded using a 100 kHz lock-in technique in a 65 T maximum field, 60 ms pulsed magnet.[22] In the first case we used the standard four-probe method with an ac resistance bridge. The temperature at high fields was controlled to ~50 mK with a typical field sweep rate of ~1 T/min by using a LakeShore capacitance censor at low temperatures, to circumvent a significant magnetoresistance of Cernox resistive sensors.

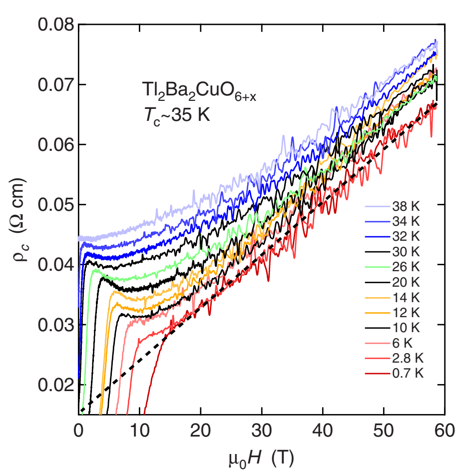

FIG. 2. (Color online) ρ_c vs magnetic field for the overdoped $Tl_2Ba_2CuO_{6+x}$ with $T_c = 35$ K. Here the peak at H_{sc} and the upturn (or negative magnetoresistance) in the $\rho_c(H)$ only becomes visible above $T = 10$ K, when the superconductivity is sufficiently suppressed by magnetic field, see also Figs. 3(a) and 3(b). Eventually, when the pseudogap closes, the upturn disappears.

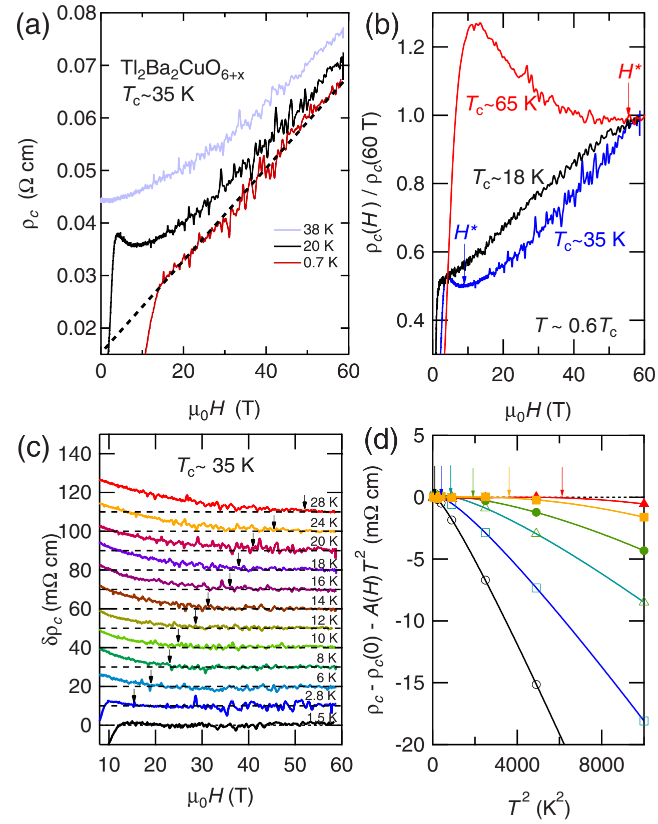

FIG. 3. (Color online) c-axis resistivity as a function of field and temperature for overdoped Tl-2201. [(a) and (b)] ρ_c vs magnetic field H at fixed temperatures exhibits a peak in the superconducting state at H_{sc}. The core feature in $\rho_c(H)$ that changes with doping is the upturn above H_{sc}—the negative MR associated with the excess resistivity due to the pseudogap. A detailed analysis of ρ_c, e.g., in Bi-2212 is in Refs. 22 and 23. For the $T_c \sim 35$ K crystal in (a) this upturn is uncovered at moderate temperatures and disappears above T_c. (b) The upturn is reduced with overdoping and is absent for $T_c \lesssim 18$ K. (c) $\delta\rho_c(H)$ obtained by subtracting the H-linear part from $\rho_c(H)$ at fixed T. Each curve is shifted vertically for clarity. $H_{FL}(T)$, marked by arrows, are the deviation points from H-linear MR. (d) ρ_c as a function of T^2 (for $T_c = 15$ K) with the T^2 contribution subtracted, for the fields 35, 30, 25, 20, 15, and 11.5 T. Arrows mark $T_{FL}(H)$ at the deviation from T^2.

III. INTERLAYER MAGNETOTRANSPORT

To probe the metallic states we used interplane charge transport. It has a number of advantages. First, c-axis resistivity is more sensitive to the pseudogap than in plane. This

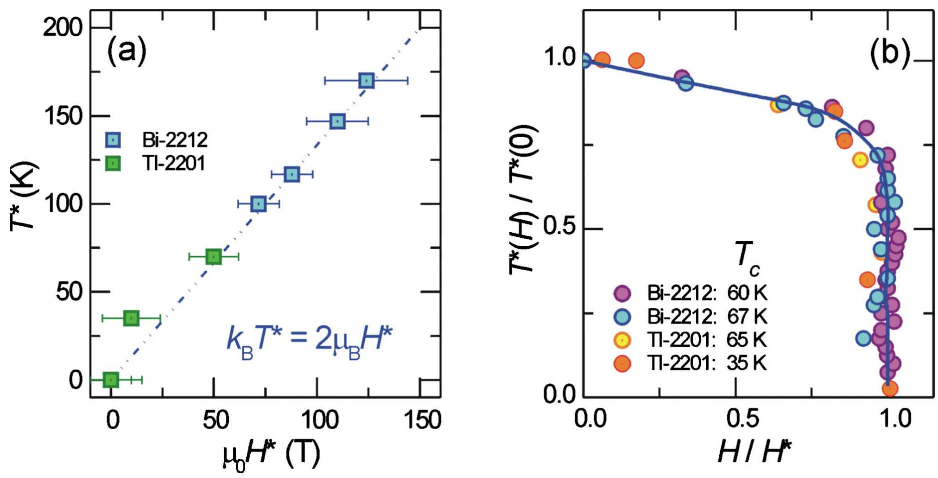

FIG. 4. (Color online) Field-temperature diagram for overdoped Tl-2201 obtained from the high-field transport measurements. (a) T_c =35 K. (b) T_c=18 K. (c) T_c=15 K. Sky blue squares, $T_{\mathrm{FL}}(H)$, and dark blue squares, $H_{\mathrm{FL}}(T)$, separate FL and n-FL states. Red open squares represent the superconducting limiting field $H_{sc}(T)$, which in cuprates varies exponentially (Ref. 22) with T. The pseudogap field H^* (brown open circles) is in evidence only for the highest T_c. The Fermi-liquid boundary is strictly linear. It terminates at $H_{\mathrm{QC}} \sim H_{c2}$, where the Fermi-liquid coefficient $A \propto \gamma^2$ in the FL [where $\rho_c(T) = \rho_c(0) + AT^2$] diverges (Ref. 21). γ is the electronic coefficient of specific heat and a measure of the effective mass m^* of a Landau quasiparticle.

was demonstrated for $Bi_2Sr_2CaCu_2O_{8+y}$ (Bi-2212),[22,23] where a semiconductinglike upturn (and the associated negative magnetoresistance, MR) in $\rho_c(T)$ above T_c—a result of the depletion in the quaisparticle density of states near Fermi energy—is a ubiquitous signature of the pseudogap. It changes systematically over a wide range of doping, consistent with the pseudogap determined by, e.g., spectroscopic techniques.[22,24,25]

Second, just as in-plane resistivity, $\rho_c(T)$ exhibits the T^2 dependence in the Fermi-liquid state and thus can be used to track the non-Fermi-liquid departures from it. Furthermore, c-axis longitudinal magnetotransport ($H \| c$) will be less affected by orbital contributions than the transverse geometry: in our heavily doped Tl-2201 we estimate a relatively low $\omega_c \tau \approx 0.4 < 1$, where ω_c is the cyclotron frequency and τ is the quasiparticle scattering time.[21]

A. Pseudogap

Figure 2 illustrates the field dependence of the c-axis resistivity ρ_c of one overdoped Tl-2201 crystal below and above its $T_c \approx 35$ K [also see Fig. 3(a)]. At the lowest temperatures the magnetoresistance in the normal state is positive and linear in field over the entire field range. As the temperature is increased above $\sim 0.5 T_c$, a small peak in $\rho_c(H)$ is articulated near the onset of superconductivity ($\rho_c \rightarrow 0$). The upturn preceding the peak—the region of negative MR—is due to filling the low-energy states by suppressing the pseudogap with magnetic field up to the field closing field H^*, see, e.g., Ref. 23. (We note in passing that negative MR cannot be generated by the orbital effects.) Above H^* the pseudogap is filled and MR becomes positive. The peak shifts to lower magnetic field and at first the upturn visibly grows with temperature. This is because with increasing temperature the superconductivity is suppressed and more of the pseudogap is exposed. At higher temperatures the negative MR is also suppressed; it disappears entirely at $T^* \gtrsim T_c$.

We remark that an identical field and temperature progression is observed in Bi-2212,[22] affirming the consistency of the pseudogap signatures in ρ_c of two different families of highly anisotropic cuprates. This is important, in view of the various pseudogap trajectories drawn.[13,26,27] T^* and H^* in both Bi-2212 and Tl-2201 follow linear dependence with doping p, and scale by a simple relation $2\mu_B H^* \cong k_B T^*(p)$, as shown in Fig. 4(a). This is distinct from the parabolic dome of $T_c(p)$ (Ref. 1) and that of the onset field of superconducting coherence, classically the upper critical field H_{c2}.[22,28]

With $T_c = 35$ K the hole concentration is relatively high and the pseudogap field $H^*(T)$ is relatively low and thus easily tracked. This should be contrasted with the less overdoped Tl-2201 ($T_c \sim 65$ K) where $H^* \gtrsim 50$ T is near the limit of the accessible field range, and with the more heavily doped crystals ($T_c \lesssim 22$ K), where the upturn becomes undetectable and at all temperatures only a positive MR is observed [Fig. 3(b)]. Thus, for low but still nonzero T_c, at hole doping levels $\gtrsim 0.258$ the pseudogap should be below the superconducting energy scale.

This experimental result can be understood if we assume (in simple mean-field theory) that the ground-state energy of the pseudogap state relative to the ungapped state (in zero magnetic field) is $\propto N(0)(T^*)^2$. Then the field that destroys the pseudogap is given by $\frac{1}{2}\chi(H^*)^2 \propto N(0)(T^*)^2$, where $\chi \approx \mu_B^2 N(0)$ is the susceptibility of the state without the pseudogap and $N(0)$ is the density of states near the chemical potential in such a state. The relation $\mu_B H^* \propto T^*(p)$ followed down to $T^*(p_{c0}) \approx 0$ is taken to define the critical point p_{c0} at $H = 0$.

An important and revealing result is displayed in Fig. 4(b). It illustrates *scaling* of $T^*(H)$ normalized to $T^*(0)$ vs field normalized to H^*, found for the two families of cuprate compounds. The scaling relation

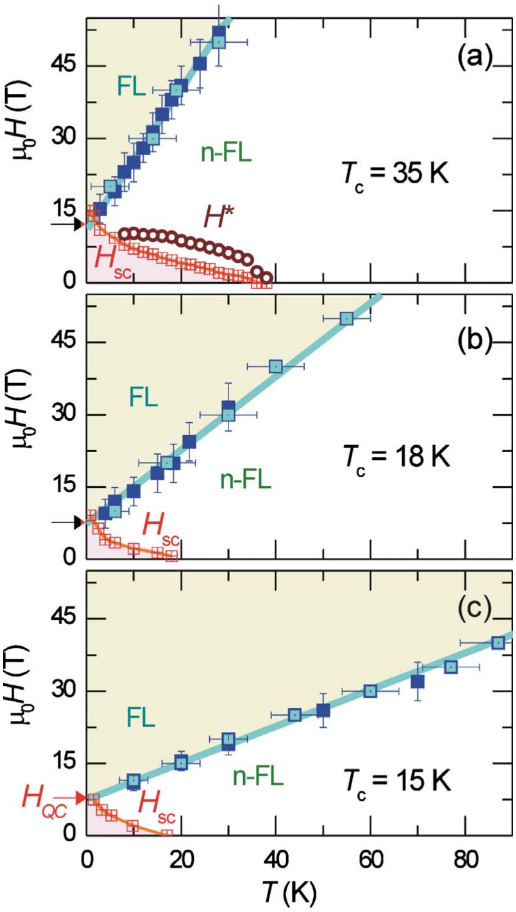

FIG. 5. (Color online) Field vs hole concentration diagram for overdoped Tl-2201 obtained from the high-field transport measurements. (a) At zero magnetic field and high temperatures, the Fermi liquid emerges beyond a nearly vertical boundary $T_{FL}(0)$ at $p \approx 0.265$, close to the edge of the superconducting dome. T^* vanishes nearby, at $p_c \sim 0.262$, but how it links with T_{FL} as $T \to 0$ is masked by the T_c dome. [Note that T^* in LSCO also vanishes well beyond optimal doping (Ref. 19).] High magnetic field, by shrinking the T_c dome, exposes the concave shape of the $T_{FL}(p, H \neq 0)$ boundary (see text), connecting with the T^* in the $T \to 0$ limit. Note that the nearly vertical onset of the $n \simeq 2$ temperature dependence seen in the in-plane transport (Fig. 1) follows a nearly vertical boundary at the edge of the dome, in close correspondence with our results. (b) Critical doping $p_c(H)$ shifts linearly from p_{c0} with field (inset), dragging the entire non-Fermi-liquid (n-FL wedge) region to lower doping. At heavy doping, it is near the edge of the superconducting state. (c) The slope of the Fermi-liquid boundary $T_{FL}(H)$ (at fixed hole concentration) diverges at $p_c \sim 0.262$. It scales with the slope of $T_{FL}(p)$ (at fixed field), see text.

$$\frac{T^*(p,H)}{T^*(p,H=0)} = \mathcal{F}\left(\frac{H}{H^*(p)}\right); \quad \mathcal{F}(0) = 1, \quad \mathcal{F}(1) = 0, \quad (1)$$

holds for different hole doping levels p. Our data demonstrates that the phase boundary at $T=0$ is vertical. From the thermodynamic[29] relation, this requires that the difference of the entropy across the transition ($\propto \partial H / \partial T$) be zero at $T=0$, consistent with the second-order quantum phase transition.[30] At finite temperatures, evidence for broken symmetry at T^* comes from the formation of nematic electronic nanostructures observed by scanning tunneling spectroscopy,[31] by the appearance of unusual magnetic order seen in neutron scattering,[17] and the spontaneous magnetic moment detected by Kerr rotation.[16]

Defining the critical point QCP by $T^*(p,H)=0$, we can consider the doping variation in $T^*(p,H)$ near $p_c(H=0) = p_{c0}$ and magnetic field variation near $H^*=0$. By taking partial derivatives, we obtain $\frac{dT^*}{dH} = (\frac{\partial T^*}{\partial p})(\frac{\partial p(H)}{\partial H})|_{p_{c0}} + (\frac{\partial T^*}{\partial H})|_{H^*} = 0$. Using experimental Zeeman scaling between $H^*(p)$ and $T^*(p)$, Eq. (1) predicts that the QCP will decrease linearly with H. Namely, $p_c(H) - p_c(0) = -(2\mu_B / k_B \alpha) H$, where $\alpha = (\frac{\partial T^*}{\partial p})|_{p_{c0}}$.

In what follows, we will examine this premise and show that indeed the FL boundary links with $T^*(p,H)=0$ to identify the QCP.

B. Conventional Fermi-liquid state

In heavily overdoped Tl-2201, as we have shown recently[21] for a sample with $T_c = 15$ K, even in the absence of the pseudogap, there is large regime at low magnetic fields where $\rho_c(T)$ does not follow a T^2-law characteristic of the Fermi liquid, but has a significant linear in T admixture.[32] (This has been also seen in the in-plane resistivity in Tl-2201 in zero field[20]). We found that the quadratic temperature dependence of resistivity is recovered in a sufficiently high field. We also found that in this Fermi-liquid MR is strictly linear in field, and the deviation from this linearity *coincides* with the deviation from the T^2 law.

Linear MR at ultrahigh magnetic fields in the Fermi liquid, while unusual at first glance, has precedents. It has been observed, for example, in $YBa_2Cu_2O_{7-\delta}$ (Ref. 33) and explained[34] as due to reduced coherence between resonant tunneling due to Landau quantization of the transverse motion in magnetic field (resembling the *quantum* linear magnetoresistance in semimetals). Field-linear MR is also obtained in the simplest class of quantum critical metals tuned by magnetic field,[35] and this effect can be very large at a field-driven QCP, at which there is the collapse of an energy scale and the field is finite.

To map the field-driven crossover to the Fermi liquid we subtract from $\rho_c(T)$ the T^2 (and the residual) term,[21] and from $\rho_c(H)$ the field-linear term, as illustrated in Figs. 3(c) and 3(d). The field-dependent crossover temperature $T_{FL}(H)$ and the temperature-dependent characteristic field $H_{FL}(T)$ obtained *independently* are indicated by the arrows. How the crossover to a Fermi liquid at $T_{FL}(H)$ boundary evolves with doping is shown in Fig. 5. Two points are evident. First, for all samples studied, the $T_{FL}(H)$ within experimental uncertainties is a simple straight line which in the zero-temperature limit tends to a finite field, that in this compound lies in the vicinity of H_{c2}, the limiting field of superconductivity. Second, the Fermi-liquid region grows rapidly with doping.

From the doping-dependent $T_{FL}(H)$ boundaries, as in Figs. 5(a)–5(c), we determine how the crossover to a Fermi liquid changes with hole concentration. The boundary in Fig.

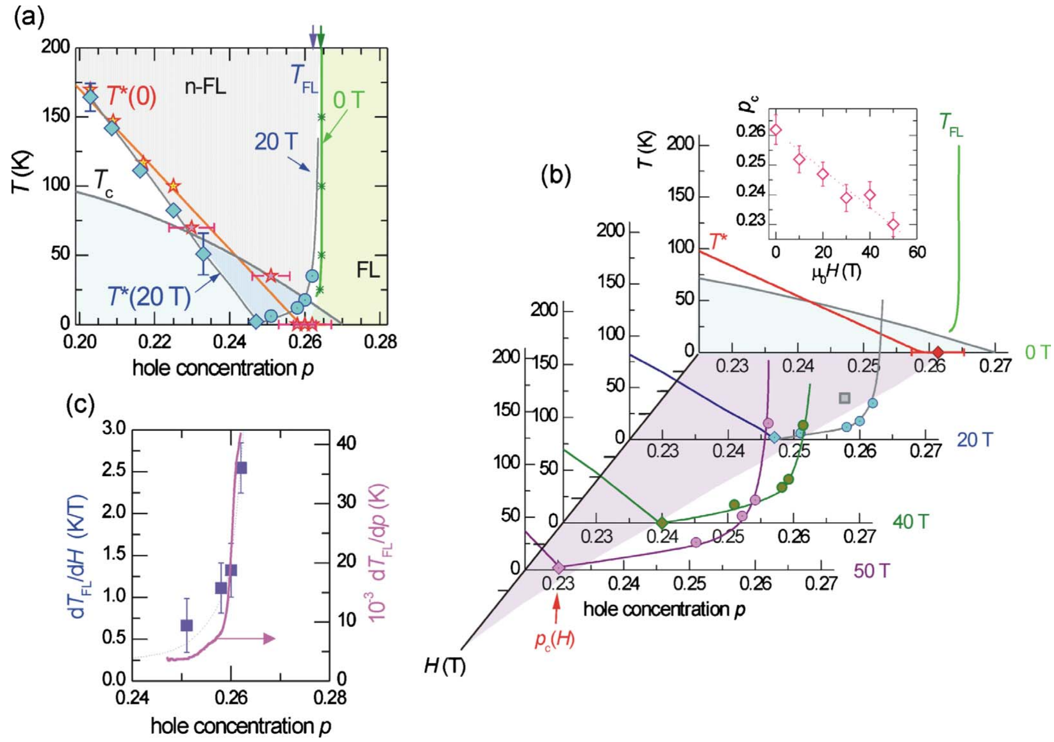

FIG. 6. (Color online) Scaling relations for the pseudogap temperature T^* observed in two families of cuprates. (a) Linear scaling relation between T^* and H^*. (b) Field dependence of $T^*(H)$ approaches H^* with a vertical slope. The slope $\partial H/\partial T$ across a phase transition is proportional to the difference of the entropy at constant H across the transition (divided by T times the change in the difference of the temperature derivative of χ). This is zero for a continuous transition at $T\rightarrow 0$.

6(a) was obtained from the $T_{FL}(H,p)$ in the H-T diagrams for different dopings and then projecting it to zero field, $T_{FL}(H\rightarrow 0,p)$. We find that at zero field and high temperatures this crossover is nearly vertical near the doping level $p\sim 0.265$. But, the quantum critical point at $H=0$ is not easily explored as we encounter the usual dilemma: at low temperatures the normal state is obscured by the T_c dome.

C. Quantum critical point

To expose the behavior of Fermi-liquid crossover at low temperatures we must suppress superconductivity (reduce the dome). This is achieved by applying a sufficiently high magnetic field. In a magnetic field, we find the boundary $T_{FL}(p,H\neq 0)$ pointing its low-temperature tail toward lower values of doping, following a concave downward path toward zero temperature. There it connects with the finite-field pseudogap line $T^*(p,H\neq 0)$, framing a "bird-beak"-shaped (asymmetric V-shaped) region that terminates at a hole concentration value $p_c(H)$. The $T^*(p)$ lines at finite fields are straightforwardly obtained from the $T^*(p,H=0)$ combined with the field dependence of $T^*(H)$ shown in Fig. 4(b).

Thus, in *toto*, in finite magnetic field the pseudogap line and the Fermi-liquid boundary meet in the low-temperature limit at a critical hole concentration level $p_c(H)$ which *linearly* decreases with increasing field [Figs. 6(a) and 6(b)]. The critical point p_c is very close to the end point of the superconducting dome (which we will discuss in the final section), and the boundaries downshift together toward lower doping side with increased magnetic field. We remark that the downslide behavior of the pseudogap critical doping value is *opposite* to the predicted field dependence of p_c arising from the spin-density-wave order competing with superconductivity in the low-doping region.[36] And we stress again that this linear decrease naturally follows from the observed critical scaling described by Eq. (1). The singular behavior at $p_c(0)=p_{c0}\approx 0.262$ is reflected in the diverging slope ($dT/dH|_{FL}\rightarrow\infty$) of the Fermi-liquid line $T_{FL}(H)$ as $p\rightarrow p_{c0}$, see Fig. 6(c).

Let us now consider $T_{FL}(p,H)$. Assuming that near the critical point the most important dependence of the FL crossover on H is through the variation in $p_c(H)$

$$T_{FL}(p,H)=T_{FL}[p-p_c(H),H]=T_{FL}\{p-p_{c0}-[p_c(H)-p_{c0}],H\}=T_{FL}(p-p_{c0}-2\mu_B/k_B\alpha H,H), \quad (2)$$

a simple relation

$$dT_{FL}(p,H)/dH|_p\approx(2\mu_0/k_B\alpha)dT_{FL}/dp|_H \quad (3)$$

in the vicinity of H^* is derived. The calculated scaling prefactor $2\mu_0/k_B\alpha\sim 0.45\times 10^3$ is very close to $\sim 0.1\times 10^3$ obtained from the data and Fig. 6(c) shows that within error bars this scaling is followed. So the singular behaviors in both, the $T^*(H)$ and $T_{FL}(p,H)$ are consistently related to the downshift of p_c.

IV. CONCLUDING REMARKS

From the interlayer magnetotransport measurements we show that the onsets of the pseudogap and the conventional Fermi-liquid state merge in the zero-temperature limit near the edge of superconductivity. We note that the crossover to the Fermi-liquid $T_{FL}(p)$ in the doping space has a very unusual shape. A form akin to this arises in a theory[10] of the critical fluctuations explaining the (orbital current) symmetry breaking in the pseudogap region.[17] The Fermi-liquid boundary when the symmetry breaking is associated with spin degrees or with unidirectional electronic (nematic) nanostructures,[31] as far as we know, has not been theoretically derived. We also note that a reanalysis of the in-plane resistivity data[20] for Tl-2201 in Fig. 1, as well as recent results in LSCO,[37] show that the onset of the T^2 temperature dependence appears to follow a similar vertical shape near the end of the dome.

It is remarkable but not unprecedented that the found QCP is pretty far from the maximum T_c—such behavior is also found in the heavy fermion system UGe_2,[38] where magnetic interactions are present. In cuprates, magnetic field can induce a static magnetic order[39] and much enhanced spin fluctuations at low T within the vortex cores.[40] The $T \rightarrow 0$ limit convergence of the pseudogap, strange metal and Fermi-liquid states at $p_c(H)$ in magnetic fields point to the direct link of the non-Fermi-liquid properties and superconductivity to the quantum critical fluctuations. Finally, critical fluctuations and anomalous behaviors reported in the spin susceptibility[20] and NMR relaxation[41] of the heavily overdoped Tl-2201 are very plausibly related.

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$=0$, where M is the uniform magnetization. However, a generalized Ehrenfest relation can be derived along the transition line: $(\frac{d\Delta S}{dT})|_{PG}+(\frac{dH}{dT})|_{PG}(\frac{d\Delta M}{dT})|_{PG}+(\frac{d^2H}{dT^2})|_{PG}\Delta M=0$. If the transition is of the second order ($\partial S=0$), then all three terms vanish separately. See, e.g., M. Roulin, A. Junod, and E. Walker, Physica C **296**, 137 (1998). Hence the vertical slope in the $T\rightarrow 0$ limit in Fig. 6(b) is consistent with the second order.

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