Quantum Phase Transition in the Magnetic-Field-Induced Normal State of Optimum-Doped High-T_c Cuprate Superconductors at Low Temperatures

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A 60 T magnetic field suppresses the superconducting transition temperature T_c in $La_{2-p}Sr_pCuO_4$ to reveal a Hall number anomaly, which develops only at temperatures below zero-field T_c and peaks at the exact location of p that maximizes T_c. The anomaly bears a striking resemblance to observations in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$, suggesting a normal-state phenomenology common to the cuprates that underlies the high-temperature superconducting phase. The peak is ascribed to a Fermi surface reconstruction at a quantum phase transition near optimum doping that is coincident with the collapse of the pseudogap state.

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High-temperature superconductivity (HTS) occurs in the transition between an undoped Mott insulator and a Fermi-liquid-like metal with a large Fermi surface (FS) [1]. Many speculate that HTS results from the proximity of a quantum phase transition (QPT) in the underlying normal state, both from the theoretical [2–6] and experimental perspective [7–11]. The existence, location, and nature of such a QPT have long been obscured by the superconducting phase; however, normal-state behavior at low temperatures emerges once the HTS phase is suppressed with intense magnetic fields [7,8], revealing a Hall effect singularity coinciding with optimal doping in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$, (BSLCO) [10], which raised a question whether the observed anomaly is a universal feature of cuprates that should be associated with the QPT.

With initial doping of the parent Mott insulator, even the question of whether the charge carriers form conventional Fermi pockets remains controversial. Angle-resolved photoemission spectroscopy (ARPES) finds well-defined quasiparticles, first near the $(\pi/2, \pi/2)$ point in the Brillouin zone, then upon further doping, extending along an arc in reciprocal space [12,13]. Debate centers on whether ARPES somehow "misses" a piece of FS: while some ARPES data suggest the Fermi arc reduces to a single point upon extrapolation to zero temperature [14], the normal-state electronic specific heat suggests the contrary [15]. Recent reports of quantum oscillations in two underdoped compounds $YBa_2Cu_3O_{6.5}$ [16] and $YBa_2Cu_4O_8$ [17,18], provide strong evidence of a small and conventional FS pocket, although its shape and location in the Brillouin zone is unknown. More recently, magnetization oscillations using the 45 T DC hybrid magnet at the National High Magnetic Field Laboratory (NHMFL) evidence a second pocket [19]. An underdoped two-pocket FS reconstruction is proposed [19], based on zone folding of a single very large FS centered on (π, π). A single very large (π, π)-centered FS has been fully mapped in overdoped $Tl_2Ba_2CuO_{6+\delta}$ from angular magneto-resistance oscillations using the same magnet [1].

The central questions remain: (a) how will the evidence for distinct underdoped and overdoped FS be reconciled, and (b) can the reconciliation provide a framework to understand both HTS and the complex phase diagram of the cuprates?

The most obvious reconciliation would be a QPT between the underdoped normal state and the overdoped normal state that is obscured by the intervening superconducting phase. Indeed, a number of HTS models propose that anomalous properties, including the linear temperature dependence of the resistivity, are governed by critical fluctuations at a QPT. Universal scaling behavior reported in neutron scattering [20], ARPES [21], and infrared spectroscopy [22] is considered as evidence of criticality. The goal of our normal-state Hall measurements is to find a hidden QPT in $La_{2-p}Sr_pCuO_4$ (LSCO), and to compare its signature with that of BSLCO [10].

LSCO thin-film samples were prepared by laser ablation using strontium titanate substrates [23] with 11 values of Sr doping, p, from 0.08 to 0.22 ($p = 0.08$, 0.12, 0.14, 0.16, 0.165, 0.17, 0.175, 0.18, 0.19, 0.20, and 0.22 with onset superconducting transition temperature, T_c, of 19.4 K, 31 K, 24 K, 28.9 K, 28.3 K, 30.2 K, 28.3 K, 27.5 K, 24.6 K, 17.9 K, and 13.6 K, respectively). All films were characterized by x-ray diffraction and checked for uniformity of low-field magnetotransport properties. All samples show metallic behavior for in-plane transport (i.e., $d\rho_{ab}/dT > 0$) at all temperatures above T_c. The samples were patterned in a conventional Hall bar geometry for measurement of the longitudinal resistivity (ρ_{ab}) and Hall resistivity ($\rho_{\rm Hall}$) using 65 T magnets at the NHMFL.

Intense magnetic fields of 50 to 65 T destroy the superconducting state to reveal the resistive normal state well below T_c. In this state, ρ_{ab} exhibits a metal to insulator (M-I) crossover [24], that occurs in our thin films at $p = 0.19$ and $\rho_{ab} \sim 0.09$ mΩ-cm, a resistivity value similar to that of LSCO and BSLCO single crystals [7,8,25].

The behavior of the $\rho_{\rm Hall}$ near optimum doping is unusual as well. Figure 1 shows (a) $\rho_{\rm Hall}$ for our $\mathrm{La_{1.86}Sr_{0.14}CuO_4}$ sample and (b) the Hall coefficient, $R_H = \rho_{\rm Hall}(H)/H$, for selected samples at 20 K. Above T_c $\rho_{\rm Hall}(H)$ is conventional, i.e., is largely linear in magnetic field for all fields [26]. Below T_c the magnetic field must suppress superconductivity before the linear-in-field normal-state behavior is recovered, denoted by the dashed lines in Fig. 1(a) that extrapolate to the origin.

The magnetic field dependence of R_H [Fig. 1(b)] has three dominant attributes: (a) there is no evidence of a magnetic-field-induced phase transition or sharp change in R_H in the normal state; (b) the p dependence of R_H in the normal state dominates the relatively small magnetic field dependence; and (c) R_H in the normal state generally becomes smaller as p is increased, the behavior of a simple metal for which the number of charge carriers is inversely proportional to the magnitude of R_H. The most striking feature of Fig. 1(b), however, is that R_H *is not a monotonic function of doping near* $p \sim 0.17$.

Figure 2 displays the temperature dependence of the high-field R_H extracted from the $\rho_{\rm Hall}$ measurements. We note that R_H is monotonic with p at high temperatures, but below 50 K, several $R_H(T)$ curves cross. Figure 2(b) magnifies our complete data set near optimum doping, evidencing a clear local minimum at low temperatures in the p dependence of R_H at $p = 0.175 +/- 0.01$.

Figure 3(a) shows the temperature and p dependence of $1/R_H$ in the normal state of LSCO. Throughout this Letter, we plot $1/R_H$ normalized to the number of holes per copper atom and refer to it as the "Hall number" ($n_{\rm Hall}$). This is a quantitatively precise notation and provides distinct advantages in communicating the magnitude of $1/R_H$ in familiar and material specific units.

For very small p, the high-temperature $n_{\rm Hall}$ has been found to be nearly equivalent to Sr doping [27] consistent with our $p = 0.08$ data in Fig. 3. In the overdoped regime, the apparent divergence of the $n_{\rm Hall}$ with increasing p is consistent with the reported zero crossing of R_H from holelike to electronlike at $p \sim 0.30$ [23,28].

The salient feature in Fig. 3(a) is the peak that develops in LSCO near $p = 0.175$ at temperatures below ~30 K. The peak (a) occurs for the values of p that gives the highest value of T_c *in this same set of samples*; (b) exhibits a narrow width of $\delta p \sim +/- 0.01$; (c) emerges only below a threshold temperature approximately equal to the maximum value of T_c; and (d) appears to have a peak value of roughly one carrier per copper atom. This peak is not unique to LSCO—a strikingly similar feature [Fig. 3(b)] with the same attributes is seen in BSLCO [10].

It is unlikely that the peak in the $n_{\rm Hall}$ can be quantitatively interpreted as the actual number of carriers, for this would imply a single-band metal with an isotropic quasiparticle scattering rate around the FS. The temperature-dependent peak, in fact, argues strongly against this sim-

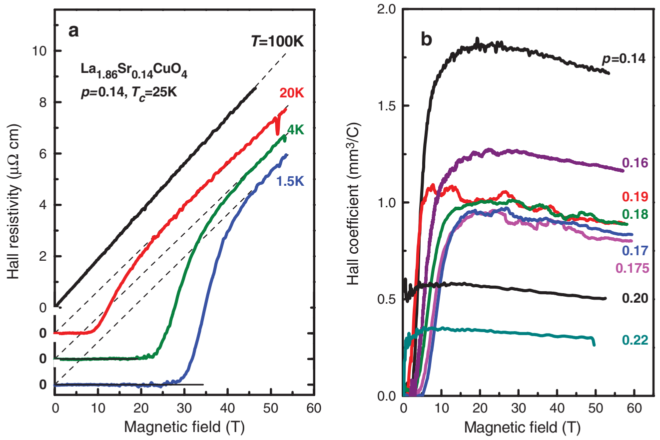

FIG. 1 (color). (a) Selected traces of $\rho_{\rm Hall}$ versus magnetic field for the $\mathrm{La_{1.86}Sr_{0.14}CuO_4}$ thin-film sample. Dashed lines are an extrapolation of the normal-state data to the origin (plots are offset for clarity). (b) R_H as a function of magnetic field at $T = 20$ K showing the nonmonotonic dependence of R_H in the normal state on doping, p. Note that the curve for $p = 0.175$ lies below the curves for *both* slightly lower doping ($p = 0.16$ and 0.17) *and* slightly higher doping ($p = 0.18$ and 0.19).

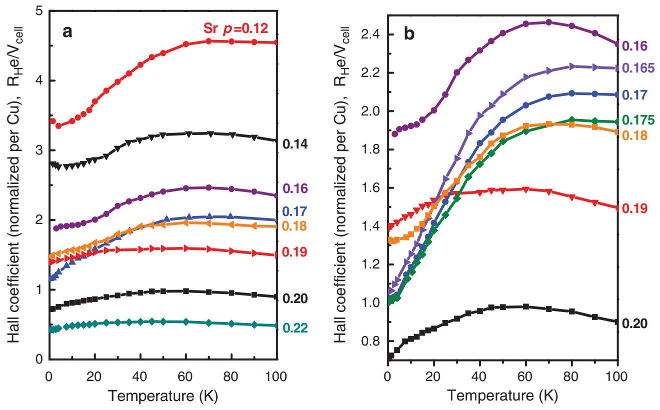

FIG. 2 (color). (a) Temperature dependence of the high-field R_H for $0.12 < p < 0.22$, normalized per Cu atom by the electric charge (e) and the unit cell volume ($V_{\rm cell}$) from the data of Fig. 1(b). For clarity, data from $p = 0.08$, 0.165, and 0.175 are not plotted. (b) An expanded view of the measurements from all samples near optimum doping. Although the high-temperature R_H is monotonic upon p, the low-temperature data show a local minimum at $p = 0.175$.

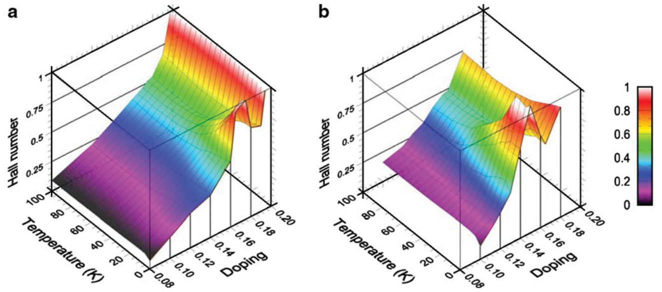

FIG. 3 (color). (a) Doping dependence of the hole-type n_{Hall} (defined as $1/R_H$ and normalized per Cu atom) in the normal state of LSCO. The grid superimposed on the data indicates the discrete data set from which the surface is deduced. The most striking feature of the n_{Hall} is the cusp at low temperatures that is centered on $p \sim 0.175$, which is the same LSCO sample in which T_c is highest. (b) n_{Hall} in platelets of BSLCO single crystals (adapted from Ref. [10]), showing a similar low-temperature peak, also occurring near optimum doping and exhibiting a similar peak amplitude near one carrier per copper atom.

plest interpretation. In Fig. 4, we subtract the n_{Hall} at 100 K from the entire data set, in effect removing the smooth background and its evolution with p in order to focus on the peak in isolation.

ARPES experiments on superconducting LSCO samples (necessarily at zero magnetic field) report FS that are holelike at $p = 0.15$ and *electronlike* at $p = 0.22$ (left and right insets in Fig. 4(c), respectively) [29]. However, similar ARPES studies performed on BSLCO argue that the FS does *not* display a change in topology anywhere in the 0.10 to 0.18 doping range [holelike FS in insets of Fig. 4(d)] [30]. Because the Hall anomaly we report near optimum doping in both LSCO and BSLCO argues for a common mechanism, we discuss our n_{Hall} peak in connection with the transition from a small-carrier-density metal, characterized by ARPES Fermi arcs, to a large-carrier-density metal with a large FS as in overdoped $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$ [1].

Although the interpretation of ARPES Fermi arcs is still debated, many have inferred a low number of quasiparticles linked to the "pseudogap" state. The development of the pseudogap has been discussed in terms of the onset of order, with many candidates for the ordered state having been proposed, including antiferromagnetic correlations [31], a d-density wave state [6], and a staggered flux phase [32]. The pseudogap state is thus often discussed in terms of a reconstruction of the FS [33,34]. Rather than enter the fray among theorists, we discuss our results largely in the context of other experimental observations.

Extrapolations of resistivity data above T_c, NMR, and specific heat data, taken together from many research groups and many values of carrier doping, suggest the collapse of the pseudogap phase at $p \sim 0.19$ [9,35]. The experimental ARPES papers also discuss the difference between Fermi arcs and the large FS in terms of the collapse of the pseudogap state because the Fermi arcs terminate in reciprocal space where there are energy gaps in the pseudogap state [12–14].

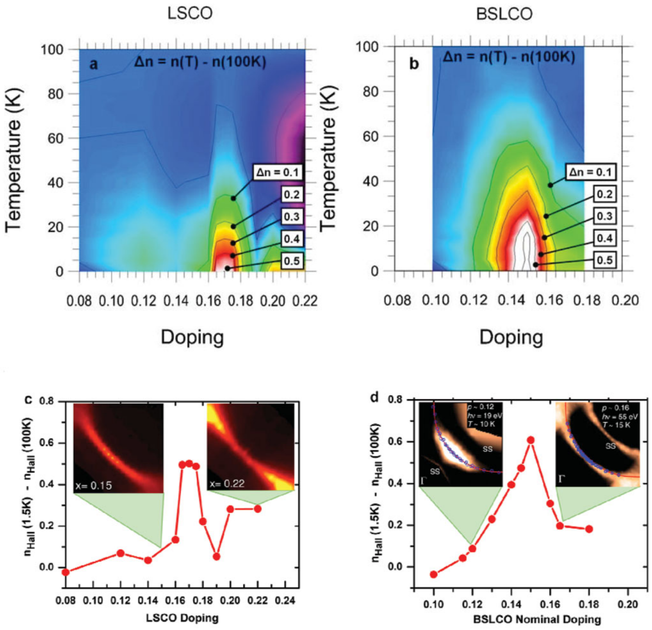

FIG. 4 (color). (a), (b) Contour plots of the Hall number variation, $\Delta n = n(T) - n(100\text{ K})$, as a function of doping and temperature in (a) LSCO and (b) BSLCO from the data of Fig. 3. (c), (d) The low-temperature ($T \sim 1.5$ K) value of the Δn versus doping in (c) LSCO and (d) BSLCO. The four insets show ARPES data for the dopings indicated, reproduced from (c) Ref. [29] and (d) Ref. [30].

How might the collapse of the pseudogap state give rise to a peak in the n_{Hall}? Whatever the specific theoretical perspective, the loss of an order parameter in the cuprates upon doping would likely be accompanied by critical fluctuations occurring within a limited doping range *and temperature range* of the quantum critical point (QCP). Those critical fluctuations have been linked to the nucleation of singular—and attractive—quasiparticle interactions [2].

Although it is beyond the present reach of theory, one can conjecture that singularly attractive interactions would delocalize electrons in the vicinity of the QCP. In this picture, the peak in the n_{Hall} would result from the delocalized electrons that would otherwise remain localized due to Mott physics. Although speculative to be sure, a link between the optimal doping for superconductivity and enhanced delocalization in the normal state might provide a natural accounting for the common temperature scale for the maximum superconducting T_c and the onset of the Hall peak anomaly in the normal state.

Regardless of whether the Hall peak is ultimately understood in terms of critical fluctuations, the observation of the same phenomena in the n_{Hall} of two different hole-doped HTS systems suggests a common quantum phase transition underlying the HTS dome. Given experimental evidence, especially from ARPES, the peak in the n_{Hall} at optimum doping is naturally interpreted as the signature of a transition from the Fermi arc state in the underdoped regime to the large FS in the overdoped regime. We note that the existence of metal-insulator crossover is common among the cuprates, but precise doping is variable, occurring in the underdoped regime in BSLCO [25], at optimum doping in LSCO single crystals [8], and in the overdoped regime in these LSCO thin films [24]. The peak in the n_{Hall}, however, occurs at optimum doping in both BSLCO and LSCO and appears to be more fundamentally linked with the optimum conditions for HTS.

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[26] A feature evident in Fig. 1(b) and not discussed in the main text is that $R_H(H)$ decreases with increasing field because $\rho_{\mathrm{Hall}}(H)$ is slightly sublinear. While this effect is sufficiently small that it does not change the salient conclusions of this Letter, we note that the departure from linearity is larger as p increases. As recently demonstrated for an electron-doped cuprate superconductor [P. Li *et al.*, Phys. Rev. Lett. **99**, 047003 (2007)], this is most likely a precursor of the eventual change of sign of R_H at $p \sim 0.3$ [23,28]. The saturation of the Hall angle, $\arctan(\rho_{\mathrm{Hall}}/\rho_{ab})$, could also contribute to nonliner Hall response at high field, particularly in the multiband scenario [19]; however, we estimate the relative effect to be of the order of $(\rho_{\mathrm{Hall}}/\rho_{ab})^2 < 2 \times 10^{-3}$ for our films.
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[35] We note that the experiments and analysis in Ref. [9] rely on measurements above T_c or on samples *that are still superconducting*, perhaps complicating comparisons to the low-temperature normal state revealed upon application of intense magnetic fields. In particular, any difference in doping levels for the two phenomena could arise due to the presence of the superconducting state or the magnetic field, or any other imperfect understanding of this “crowded” region of phase space in the vicinity of an ill-understood quantum phase transition.