Evidence for a Quantum Phase Transition in $Pr_{2-x}Ce_xCuO_{4-\delta}$ from Transport Measurements

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The doping and temperature dependences of the Hall coefficient, R_H, and *ab*-plane resistivity in the normal state down to 350 mK is reported for oriented films of the electron-doped high-T_c superconductor $Pr_{2-x}Ce_xCuO_{4-\delta}$. The doping dependences of β ($\rho = \rho_0 + AT^\beta$) and R_H (at 350 mK) suggest a quantum phase transition at a critical doping near $x = 0.165$.

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The mechanism for the superconductivity (SC) in the high-T_c copper oxides is still a mystery in spite of 17 years of intense research on these materials [1]. Moreover, many of the normal state properties are not understood either. Of the many ideas put forth to explain the cuprates, the existence of a quantum phase transition (QPT) between two phases near a critical doping, x_c, and its relationship to the unconventional SC is emerging as an important issue [2–4]. The nature of these phases and their relation to the origin of the high-T_c SC is unclear at present. Although a QPT occurs at $T = 0$, it is characterized by a "funnel-shaped" region in the p-T phase diagram, where p is the control parameter [5]. The physical properties in this region are governed by quantum fluctuations. A study of some of these physical properties in the electron-doped cuprates, with the doping level being the control parameter, is the focus of the research presented in this Letter.

Thus far, the evidence for a QPT in the hole-doped (p-type) cuprates has been somewhat indirect [6] because the upper critical fields (H_{c2}) are extremely large, and it is not possible to destroy the SC to examine the normal state properties down to very low temperatures. A recent exception [7] will be discussed later. Because of the lower H_{c2} (10 T maximum at optimal doping), the electron-doped cuprates ($R_{2-x}Ce_xCuO_{4+\delta}$ with R = Nd, Pr, La, Sm) offer a distinct advantage for investigating the consequences of a doping dependent QPT in the cuprates. In the electron-doped (n-type) cuprates, an antiferromagnetic (AFM) phase starts at $x = 0$ and persists up to, or into, the SC dome [8]. Recent neutron scattering experiments [9] show an AFM phase for $H > H_{c2}$ in optimally doped n-type cuprates, but no such phase on the overdoped side. These experiments suggest that AFM is the competing phase with SC in the n-type cuprates. Tunneling measurements give evidence for a normal state pseudogap for dopings within the SC dome [10] which goes away near $x = 0.17$ [11]. It is not known if this pseudogap is similar to that found in the p-type cuprates or if it is related to the AFM order. Angle-resolved photoemission spectroscopy (ARPES) experiments on $Nd_{2-x}Ce_xCuO_{4-\delta}$ show that the Fermi surface (FS) changes dramatically as a function of doping, going from a small electron pocket at $(\pi, 0)$ in underdoped ($x = 0.04$) to a large holelike FS at optimal doping ($x = 0.15$) [12]. Neither the magnetic measurements nor ARPES have had the energy or doping precision to determine x_c. The detailed low temperature transport experiments presented here show that a quantum critical point (QCP) occurs at $x_c = 0.165$ (± 0.005) in $Pr_{2-x}Ce_xCuO_{4-\delta}$ (PCCO).

The normal state transport properties of the cuprates for $T > T_c$ do not follow the behavior expected for conventional metals [Fermi liquid (FL)]: the Hall coefficient has a strong T dependence, the *ab*-plane resistivity varies as $\rho \propto T$ (for p-type), $\rho \propto T^2$ (for n-type), up to temperatures greater than 250 K. A marginal FL model has been successful in explaining these properties in the hole-doped cuprates (as well as some other properties) [13]. This model is compatible with the notion of a QPT near optimal doping [3]. In various QCP models, there are definite predictions for the behavior of the resistivity and the Hall effect near the QCP [13,14]. For example, in a d-density wave picture, a sharp kink in R_H as a function of doping has been predicted [15]. Evidence for some of these predictions has been found in other correlated systems such as heavy fermions [16]. In the cuprates, other suggestive evidence for a QPT under the SC dome has come from the observation of a low T normal state "insulator" to metal crossover as a function of doping in both n- and p-type materials [17,18].

In this Letter, we present a comprehensive study of the *ab*-plane resistivity and Hall effect in films of electron-doped PCCO, for many dopings near and within the SC dome. We study the normal state ($H > H_{c2}$) from 350 mK to 300 K with an emphasis on the temperature and doping dependence of these transport properties below 20 K. Our results give compelling evidence for a QPT at a critical doping of $x = 0.165 \pm 0.005$ (i.e., in the overdoped region of the phase diagram).

PCCO c-axis oriented films of various Ce doping concentrations were deposited from stoichiometric targets on (100) oriented $SrTiO_3$ substrates using the pulsed laser deposition (PLD) technique with conditions similar to those reported by Maiser *et al.* [19]. Rutherford backscattering measurements were used to determine the

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thickness of the films. The minimum-channeling yield obtained was 10%–20% indicating a good epitaxial growth. The low residual resistivity and sharp T_c (see Fig. 1) indicate that the films are of better quality than the best previously reported PLD films [18] and comparable to molecular-beam epitaxy grown films [20]. Since the oxygen content has an influence on both the SC and normal state properties of the material [21], we took extra care in optimizing the annealing process for each Ce concentration. We found that the optimal annealing time, t_A, after which T_c stopped changing, increases with increasing Ce doping. We also found that roughly $t_A \propto d^2$, with d being the films' thickness, as expected for a diffusion process. For $x \leq 0.15$, we had to simply maximize T_c and avoid decomposition spots detectable in an optical microscope.

Resistivity and Hall voltage were measured up to a magnetic field of 14 T and down to a temperature of 0.35 K with the field applied perpendicular to the CuO_2 planes. At low temperatures, the Hall coefficient was measured by taking field scans from -14 to 14 T, a field at which the PCCO is well in the normal state even at 0.35 K.

In Fig. 1, we show the *ab*-plane resistivity versus temperature at 10 T ($H > H_{c2}$) for $0.13 \leq x \leq 0.19$. First, we note the decrease in the resistivity as the Ce concentration is increased. Another feature appearing in $x \leq 0.15$ films is a sign change in $d\rho/dT$ (an upturn). The temperature at which the upturn appears decreases with increasing doping. The inset shows the resistive SC transition where T_c has the expected doping dependence [19]. All the films have sharp transitions. The transition width, ΔT_c, measured as the width at half maximum of the peak in $d\rho/dT$, is $\Delta T_c = 0.3$–0.6 K in optimum and overdoped PCCO (increasing with increasing Ce doping), and $\Delta T_c = 2.2$ K for $x = 0.13$, all much sharper than previously reported PLD films.

Figure 2(a) shows typical Hall resistivity ρ_{xy} at various temperatures for the $x = 0.17$ sample. Note that ρ_{xy} is linear as a function of magnetic field above the upper critical field ($H_{c2} \approx 5$ T for $x = 0.17$). The normal state resistance recovers well below 10 T; hence, the measurements taken at high fields are normal state measurements. We calculate the Hall coefficient, R_H, from the slope of a linear fit to the data at high fields ($\rho_{xy} = R_H B$). In Fig. 2(b), we show R_H as a function of temperature for various doping levels. Note the strong temperature dependence in the intermediate doping levels even at $T < 10$ K. The sign of R_H, which in a simple metal corresponds to the type of charge carrier, also changes with doping and temperature. The temperature dependence of R_H (along with other transport properties) was previously interpreted as evidence for two types of carriers for Ce concentrations near optimal ($x = 0.15$)

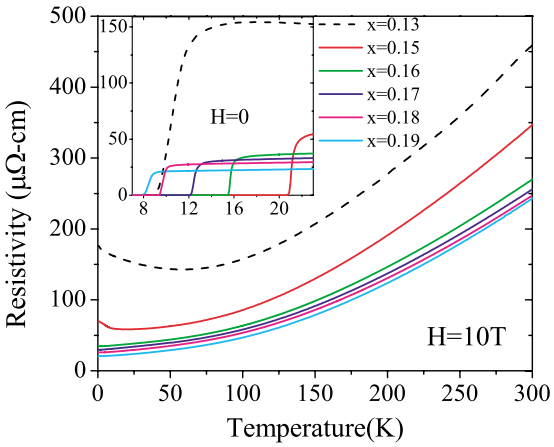

FIG. 1 (color online). *ab*-plane resistivity versus temperature for $Pr_{2-x}Ce_xCuO_{4-\delta}$ films with various Ce doping (x) in a magnetic field of 10 T ($H > H_{c2}$) applied parallel to the c axis. Inset: Resistive superconducting transition $H = 0$.

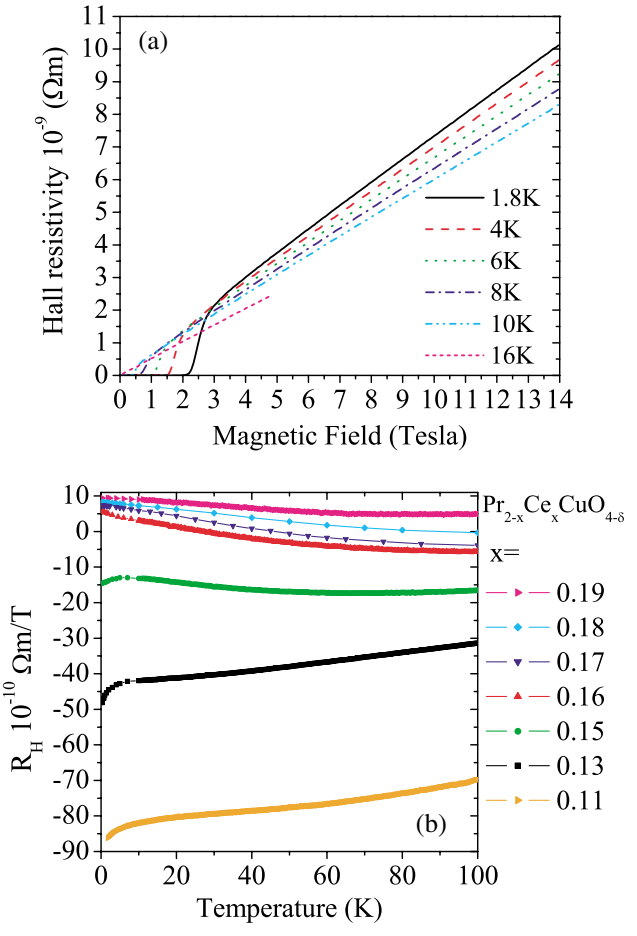

FIG. 2 (color online). (a) Hall resistivity (ρ_{xy}) for the PCCO $x = 0.17$ film. The Hall coefficient, R_H, is the slope of a least square fit from well above the upper critical field to 14 T for $T < T_c$, and from 0 to the highest field measured, at $T > T_c$. (b) The Hall coefficient as a function of temperature for the various Ce doping.

[22]. Although it is not possible to determine the exact oxygen content in the films, R_H at low temperatures depends systematically on Ce doping, which suggests using the low temperatures R_H as a criterion to determine the number of carriers (coming from both Ce and oxygen).

In Fig. 3 we show R_H at 0.35 K as a function of cerium doping for $x = 0.11$ to $x = 0.19$. An abrupt change in R_H is seen above $x = 0.16$. This is one of the important new results of our work. Previous studies did not measure enough samples to reveal this striking change in R_H. R_H at low temperatures, being free from complicated inelastic scattering processes, reflects the electronic structure of the material. Therefore, the abrupt change in R_H at 0.35 K is an indication of a significant reorganization of the FS, which we believe may result from a QPT between two phases in the normal state. From the R_H behavior at 0.35 K, we identify one phase at low x where R_H changes rapidly and another phase at high x where R_H varies more slowly. We find the QCP at $x_c = 0.165 \pm 0.005$ from the intersection of best fit straight lines through the high x and low x data. This is just above the doping where R_H crosses zero (somewhere between 0.16 and 0.15), a fact that can be determined only from the lowest T data. The QCP that we precisely determine from R_H (and below from resistivity) is consistent with the doping trends seen in the magnetic [8], tunneling [11], and ARPES [12] measurements.

Now we identify in our resistivity measurements the QCP found in the Hall data. We fit the important low temperature range (0.35 to 20 K) of the resistivity data from Fig. 1 to the form $\rho(T) = \rho_0 + CT^{\beta}$, with C, ρ_0, and β independent of temperature. As an example, $\rho(T)$ for $x = 0.17$ and the fit are shown in Fig. 4(a). The exponent, β, obtained from the fits is presented in Fig. 4(b). It has a strong doping dependence and gets closer to 1 as we decrease the Ce doping from 0.19 to 0.17. Decreasing the Ce doping further to $x = 0.16$ results in an increase in β to 1.4. One should note that at high temperatures ($T > 30$ K) the resistivity follows a T^2 behavior as was previously reported [18]. At very low temperatures, the temperature dependence is again T^2. The T^2 region starts below $T_0 = 4.8$, 4.7, 2, and 6.8 K $\pm 5\%$ in $x = 0.19$, 0.18, 0.17, and 0.16, respectively (for $x \leq 0.15$ the low temperature behavior is obscured by the upturn). Between the two T^2 regions, we find the different temperature dependence with exponent $1 < \beta < 2$. By fitting our data over a finite temperature range (0.35–20 K), we are averaging over a Fermi liquidlike $\rho \propto T^2$ region at the lowest temperatures (below T_0) and a quantum critical region with $\beta < 2$. Thus, we obtain a fractional exponent which gets closer to 1 for x approaching x_c and which goes back up towards 2 as x gets greater than x_c. Fournier *et al.* [18] found a linear in T resistivity from 10 K down to 40 mK in one of their Ce = 0.17 PCCO films, a film that had $T_c = 15 \pm 4$ K, somewhere between our $x = 0.16$ and $x = 0.17$ samples. It is possible that Fournier *et al.* hit x_c in their Ce = 0.17 film. Note that x_c depends on both Ce and oxygen, and therefore samples made by different groups can differ slightly in Ce concentration for the

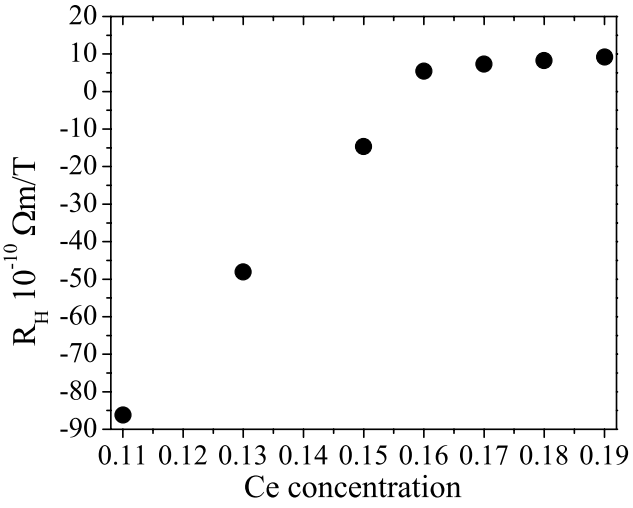

FIG. 3. The Hall coefficient at 0.35 K [taken from Fig. 2(b)]. A distinct kink in the Hall coefficient is seen between $x = 0.16$ and$x = 0.17$. The error on the concentration is approximately 0.003. The error in R_H comes primarily from the error in the film thickness; it is approximately of the size of the data points.

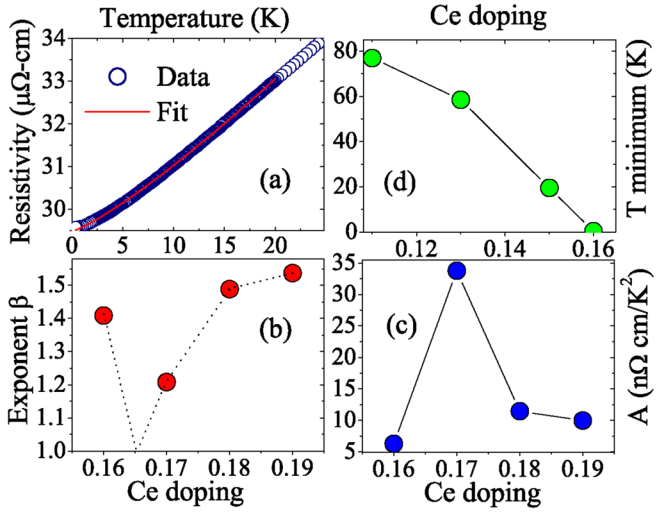

FIG. 4 (color online). (a) Analysis of the data from Fig. 1. We fit the data from 0.35 to 20 K to $\rho = \rho_0 + AT^{\beta}$: circles, the $x = 0.17$ film data; solid line, fit. (b) The doping dependence of the exponent β found from a fit from the lowest measured temperature up to 20 K. The error bars are smaller than the symbols. Lines are guides to the eye. (c) The coefficient A in a fit to the form $\rho = \rho_0 + AT^2$ in the low temperature T^2 region (described in the text) as a function of doping. (d) The temperature at which minimum resistivity occurs for $x \leq 0.15$ as a function of doping. The resistive upturn is not found above 350 mK for $x \geq 0.16$.

same carrier concentration. Also, as shown by Rosch [14], disorder can affect the exponent in the QCP region. Based on the value of the residual resistivity, the films of Fournier *et al.* have different disorder than ours. Taking Fournier's data into account, β appears to approach 1 around $x_c = 0.165$ in our films, the same doping at which we found the kink in R_H. Thus, Fig. 4(b) suggests that in a "funnel-shape" region in the doping-temperature phase diagram a linear, or close to linear, in T resistivity occurs; this funnel ends in a unique doping level, x_c at $T = 0$ K. This is the behavior expected for transport properties in the quantum critical fluctuation region at finite temperatures above a QCP [2–5].

To give further support to the QPT scenario, in Fig. 4(c) we plot the coefficient A obtained when we fit the data to the form $\rho = \rho_0 + AT^2$ in the low temperature T^2 region, below T_0, as a function of doping. From the continuity of the resistivity, A should diverge as one approaches a QCP. We find a large increase in A for $x = 0.17$, the doping at which β has its smallest value and very close to the point where R_H changes abruptly. Finally, we note that the temperature of minimum resistivity where an upturn appears in $\rho(T)$ behaves in a similar way [Fig. 4(d)]; it decreases with increasing doping and vanishes on the overdoped side.

Our results are very similar to those found in heavy fermion materials where the resistivity has a power law dependence in temperature and this power law changes significantly with a control parameter p as p is varied across the quantum critical region [16,23]. Along with the abrupt R_H change at $x_c = 0.165$ (Fig. 3), the power law variation in resistivity approaching 1 at the same x_c is strong evidence for a QCP at x_c.

Note that the exponent $\beta = 1$ is expected in the marginal Fermi liquid phenomenology [13]. It is also generic that near a 2D QCP inelastic processes scale with T [2,24]. Subtle effects associated with transport present theoretical complications that at very low temperature and in very clean systems predict other powers in T [25]. However, other calculations involving systems with finite disorder [14], and the experiments in the heavy fermion systems, show that these complications are generically not significant in the accessible temperature regimes and the quantum critical predictions for β are found [16].

Very recently, Balakirev *et al.* [7] reported an abrupt change in Hall number near optimum doping in hole-doped $Bi_2Sr_{1-x}La_xCuO_{6+\delta}$, which they associated with a change in the FS associated with a QPT. However, no evidence from ARPES experiments were found for a such FS reorganization. In another hole-doped system, $La_{2-x}Sr_xCuO_4$, the FS changes from holelike to electronlike at much higher doping [26].

In summary, we have presented low temperature, normal state ($H > H_{c2}$) resistivity and Hall effect data as a function of doping, which gives compelling evidence for a quantum phase transition in the electron-doped cuprate $Pr_{2-x}Ce_xCuO_{4-\delta}$. The Hall coefficient at low temperatures shows a kink near $x = 0.165$ that suggests an abrupt change in the Fermi surface near that doping. The low temperature resistivity shows a temperature dependence with an exponent which is getting closer to 1 as one approaches $x_c = 0.165 \pm 0.05$ from the overdoped or from the underdoped side. The coefficient A, found from a fit of the lowest temperature region to $\rho = \rho_0 + AT^2$, increases by a factor of 3 at $x = 0.17$ compared to $x = 0.16$ and $x > 0.17$. The upturn in the *ab*-plane resistivity vanishes around $x = 0.16$. All of these findings strongly suggest a quantum phase transition near $x = 0.165$. The nature of the QPT cannot be determined from our transport measurements but other evidence for antiferromagnetic order suggests that in the *n*-doped cuprates the QPT is a magnetic one.

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