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Insulator-Metal Crossover near Optimal Doping in $Pr_{2-x}Ce_xCuO_4$: Anomalous Normal-State Low Temperature Resistivity

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Normal-state resistivity measurements at high fields and low temperatures in electron-doped $Pr_{2-x}Ce_xCuO_4$ thin films reveal an insulator-metal crossover near a doping level $x \approx 0.15$, similar to a previous report on hole-doped $La_{2-x}Sr_xCuO_4$. The temperature dependence of the resistivity of insulatinglike samples is sublogarithmic, while for metallic samples (with $x = 0.17$) the resistivity is linear from 40 mK to 40 K. This surprising latter observation suggests an unusual contribution to the scattering processes at low temperature in these materials. We conclude that the ground state at $x = 0.15$, corresponding to the maximum transition temperature, is equivalent for hole- and electron-doped cuprates. [S0031-9007(98)07689-3]

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Electron-doped cuprate superconductors [1] (i.e., $R_{2-x}Ce_xCuO_{4+\delta}$, with R = Nd, Sm, Pr) represent an important challenge to any theory attempting to describe the mechanism of superconductivity in the high temperature superconducting cuprates. Because they share with the hole-doped cuprates a common building block, namely, the copper-oxygen plane, one expects the same mechanism to apply. The appearance of superconductivity for both types of carrier doping might suggest an electron-hole doping symmetry [2]. However, this proposed symmetry is questionable since many properties have been found to be different. First of all, the critical temperature T_c is strongly peaked between $x \approx 0.12$ to 0.18, in contrast to the wider, bell-shaped, x dependence found in $La_{2-x}Sr_xCuO_4$ (LSCO). Second, the ab-plane and c-axis resistivities, respectively, ρ_{ab} and ρ_c, of optimally doped ($x = 0.15$) thin films and single crystals follows roughly a T^2 dependence [3,4] rather than $\rho_{ab} \propto T$ and insulatinglike ρ_c found in the optimal hole-doped cuprates ($x = 0.15$ for LSCO). This difference has been suggested as evidence that the electron-doped cuprates are *overdoped* [5], since the normal-state and superconducting properties are comparable to overdoped LSCO (with $x \geq 0.20$). Finally, optimally doped $Nd_{2-x}Ce_xCuO_{4+\delta}$ has a penetration depth [6] and Raman scattering [7] consistent with an s-wave superconducting order parameter.

Probing the normal-state transport properties of the copper oxide superconductors down to the lowest temperatures (toward the ground state of the system at $T = 0$), for the entire doping range, is important in order to provide information on the anomalous nature of the normal state [8]. However, because of the T_c values and the corresponding very large upper critical field (H_{c2}), studies of the normal-state properties for $T < T_c$ have been limited. To reach the normal state below T_c, one must apply a magnetic field larger than the upper critical field (H_{c2}). Ando, Boebinger, and co-workers [9–13] have used pulsed magnetic fields up to 60 T to several hole-doped cuprates with low T_c. They found a low temperature insulator-metal crossover as a function of doping close to optimal doping ($x = 0.15$) for $La_{2-x}Sr_xCuO_4$ in Ref. [11]. The same authors showed that this crossover occurs for a value for the copper-oxygen plane sheet resistance of $R_{sh} \approx (1/12)(h/e^2)$ ($R_{sh} \equiv \rho_{ab}/c$), corresponding to an uncharacteristic large value of $k_F l \approx 13$, thus suggesting an anomalous nature for the normal-state properties.

In this Letter, we demonstrate that a similar insulator-metal crossover is found in electron-doped $Pr_{2-x}Ce_xCuO_{4+\delta}$ thin films. It occurs close to optimal doping ($x = 0.15$) at an anomalously small sheet resistance corresponding to $k_F l$ between 20 and 25. Thus, despite the numerous disparities mentioned above, the ground state ($T = 0$) of the CuO_2 planes appears to be independent of the carrier type, as an insulator-metal crossover at near optimal doping is observed for both hole- and electron-doped superconductors. We also observe an unusual linear temperature dependence of the resistivity down to 40 mK for $x = 0.17$. This is interpreted as a strong evidence of electron-electron inelastic scattering in the normal state of the overdoped electron-doped cuprates.

We chose to measure the transport properties on c-axis oriented epitaxial thin films of $Pr_{2-x}Ce_xCuO_4$ (PCCO) prepared by pulsed-laser deposition [14,15], since the growth of electron-doped crystals has often been plagued by Ce inhomogeneity problems [16]. In a previous detailed study [14], the films were shown to reproduce closely the target composition, thus allowing the better compositional control necessary for our present study. They were patterned into small Hall bars with thicknesses of 3000 to 8000 Å allowing simultaneous measurements of the resistivity and the Hall effect [17]. The films

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studied here have an optimal oxygen content obtained by maximizing T_c for a given Ce content. The results were reproducible for each Ce composition.

Figure 1 shows the resistivity at 0, 8.7, and 12 T as a function of temperature for the PCCO thin films in the composition range which exhibit superconductivity at $H = 0$. At higher temperature, all of the films show a temperature dependence close to T^2 above T_c (not shown here [17]), as found previously [3]. Moreover, the maximum upper critical field (H_{c2}) at $T = 0$ is of the order of 10 T [17]. The low temperature resistivity at high fields varies very little below T_c. However, a resistivity minimum and an upturn toward insulatorlike behavior for $x = 0.13$, 0.14, and 0.15 can be clearly detected in Figs. 1(a) and 1(b). This is not the case for the $x = 0.17$ and 0.20 films, which show metallic resistivity down to the lowest temperatures measured [18]. Actually, the minimum in resistivity for the $x = 0.15$ films occurs below T_c ($T_{\min} \approx 15$ K), a result very similar to the one reported for optimally doped $La_{2-x}Sr_xCuO_4$ [11] and La-doped $Bi_2Sr_2CuO_6$ [9] crystals.

In Figure 2, the resistivity at 12 T for the films with $H_{c2}(0)$ close to 8.7 T (i.e., $x = 0.14$, 0.15, and 0.17) is magnified to show the details of their temperature dependence. The measurements down to 40 mK clearly show that the resistivity continues to increase steadily as the temperature decreases for $x = 0.14$ [Fig. 2(b)] and $x = 0.15$ [Fig. 2(d)] showing no indication of downward curvature (from onset of superconductivity), while ρ_{ab} decreases linearly for $x = 0.17$ [Fig. 2(e)]. Our low temperature data confirm the nonmetal (or "insulator" with $\partial\rho_{ab}/\partial T < 0$) to metal ($\partial\rho_{ab}/\partial T > 0$) crossover near optimum doping for the ground state ($T = 0$) of the copper-oxygen planes with electron (cerium) doping. We should emphasize that the resistivity for $x = 0.14$ and 0.15, over almost three decades of temperature shown in Figs. 2(a) and 2(c), is *not* following a simple $\rho_{ab} \propto \log(1/T)$ dependence as is expected for two-dimensional (2D) weak localization [19,20]. Our sublogarithmic trend is in contrast with the $\log(1/T)$ behavior reported for underdoped LSCO [12], and remains a puzzling fact. One possible explanation is our extended temperature range of measurements compared to the LSCO data [11], allowing us to see deviations from the logarithmic behavior at low temperatures. Possible deviations due to large (negative) magnetoresistance cannot be ruled out. Another possibility is the very narrow range of cerium doping which corresponds to underdoping in PCCO, i.e., from $x = 0.12$ to 0.15, which does not allow for a clear comparison with underdoped LSCO. It appears that the variation of ρ_{ab} in the "insulating" temperature range is much stronger in LSCO than in PCCO for equivalent doping: for example, at $x = 0.15$, ρ_{ab} for LSCO doubles from 30 to 0.65 K (Fig. 1 of Ref. [11]), while it increases barely by 10% for PCCO. This difference might indicate that localization effects are (somehow) weaker in PCCO than LSCO. Interestingly, La-doped $Bi_2Sr_2CuO_y$ close to optimal doping behaves similarly [9] to our PCCO thin films. Other temperature dependences [for example, due to variable-range hopping (VRH), as $\exp(-T_o/T)^n$ with $n = 1/2$, $1/3$, and $1/4$] have been explored, but none reproduce the data convincingly. This is not surprising since VRH should be observed only when $k_F l < 1$ [21,22], which is clearly not the case for optimally doped PCCO with estimated values of $k_F l \approx 20$ (see below). We conclude

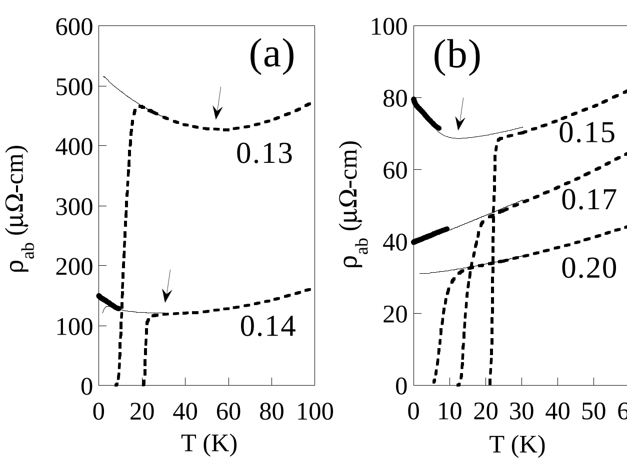

FIG. 1. Resistivity ρ_{ab} as a function of temperature for the c-axis oriented $Pr_{2-x}Ce_xCuO_4$ thin films in magnetic fields of 0 T (dashed lines), 8.7 T (thin lines), and 12 T (thick lines). (a) $x = 0.13$ and 0.14; (b) $x \geq 0.15$. The field is applied along the c axis.

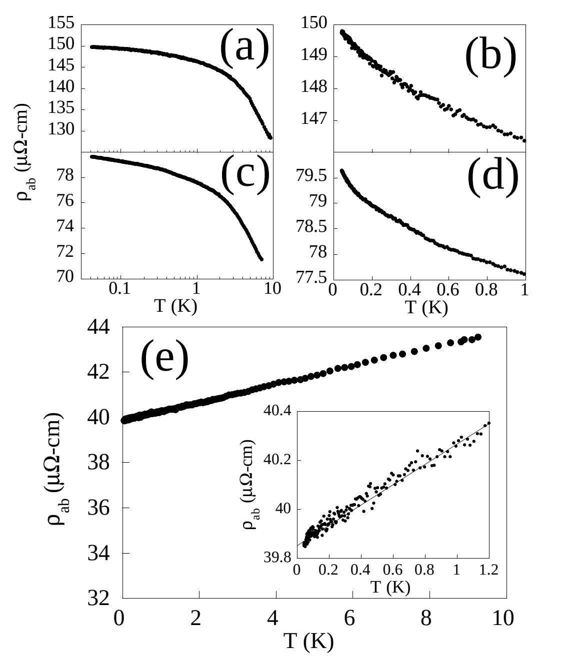

FIG. 2. ρ_{ab} below 10 K and at 12 T for some samples of Fig. 1: (a) and (b) $x = 0.14$; (c) and (d) $x = 0.15$; and (e) $x = 0.17$. In (e), the inset shows a magnified view of the subkelvin range.

that a more complete picture of the insulating state should involve the measurement of other properties, as was reported recently with the Hall effect of LSCO crystals and thin films [13]. These measurements are in progress on our samples.

As an arbitrary limit between the metallic and insulating phases, we can choose the temperature, $T_{\min}$, corresponding to the minimum in ρ_{ab} in the normal state. This is shown in Fig. 3 as a function of cerium content. This "phase diagram," very similar to the one found for LSCO [11], clearly shows that the $T = 0$ insulating phase vanishes also at a concentration close to $x = 0.15$ corresponding to the maximum T_c of the system (optimal doping). Thus, although there are differences in many properties between the electron- and hole-doped single-layer cuprates, the doping dependence of the $T = 0$ normal state appears to be similar. This suggests that optimally doped PCCO *is not in the overdoped regime* [5] and that its ground state is equivalent to LSCO $x = 0.15$. To reach a definitive conclusion on the equivalence of the crossover in PCCO and LSCO, more experiments are needed on the real nature of the nonmetallic and metallic phases and the link to the actual carrier concentration. The insulator-metal crossover occurs at a sheet resistance of $R_{\text{sh}} \approx 1000-1300\ \Omega/\square$. This range of values corresponds to an anomalously large value of $k_F l \approx$ 20 to 25 (using a cylindrical Fermi surface giving $k_F l = hc_o/\rho_{ab} e^2$). As suggested in Ref. [11], a large $k_F l$ at the crossover might be a result of the non-Fermi-liquid nature of the cuprates. As the cerium content increases, one expects the impurity scattering term to increase steadily. The decreasing residual resistivity observed in both our PCCO thin films and LSCO samples [10–12] clearly indicates that carrier concentration dominates the doping dependence.

Two striking features of our new data are the absence of a low temperature saturation of the metallic resistivity for the slightly overdoped $x = 0.17$ samples and the corresponding linear-T resistivity from 30 K down to 40 mK [see Fig. 2(e) and its inset]. Since electron-phonon scattering should be ineffective in the subkelvin range, the temperature-dependent contribution to the scattering is probably of electronic nature. A crossover of the temperature dependence from linear to quadratic is observed near 40 K. Interestingly, the same kind of linear-T dependence (and the crossover to T^2) has been observed in strongly overdoped $Tl_2Ba_2CuO_{6+\delta}$ (TBCO) crystals (with $T_c \approx 11\ K$), where a linear term persists down to 100 mK [23,24] when a large magnetic field is applied. This would suggest a similar ground state for slightly overdoped PCCO $x = 0.17$ and strongly overdoped TBCO.

It was suggested that the near-T^2 high temperature resistivity of the electron-doped cuprates could be the indication of electron-electron scattering [3,25]. However, the temperature dependence was found to deviate from the quadratic behavior at high temperatures. Tsuei *et al.* [3] showed that the resistivity can be fit to $T^2 \ln(T_F/T)$ suggesting that the resistivity is actually consistent with Fermi-liquid theories of electron-electron scattering in pure 2D metals [26,27]. However, the prefactor obtained by such a fit is 2 to 3 orders of magnitude larger than theoretical predictions [26,27]. Moreover, as we have seen in Figs. 1 and 2, the resistivity of this $x = 0.17$ film is *linear* at low temperature, another discrepancy with respect to the $T^2 \ln(T_F/T)$ predictions. To understand the origin of this crossover from T^2 to T temperature dependence, we show the resistivity and the Hall coefficient of the $x = 0.17$ film in Fig. 4, where the arrow indicates approximately where the resistivity crosses over from linear to quadratic. We observe that the linear resistivity

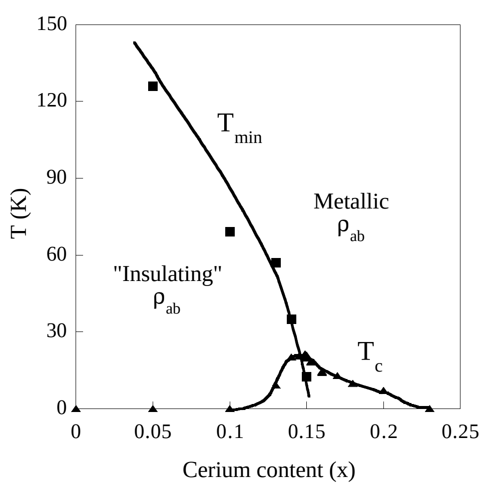

FIG. 3. Phase diagram determined from resistivity data of $Pr_{2-x}Ce_xCuO_4$ thin films. Solid triangles and solid squares are T_c and $T_{\min}$ (defined in text), respectively. The solid lines are guides to the eye.

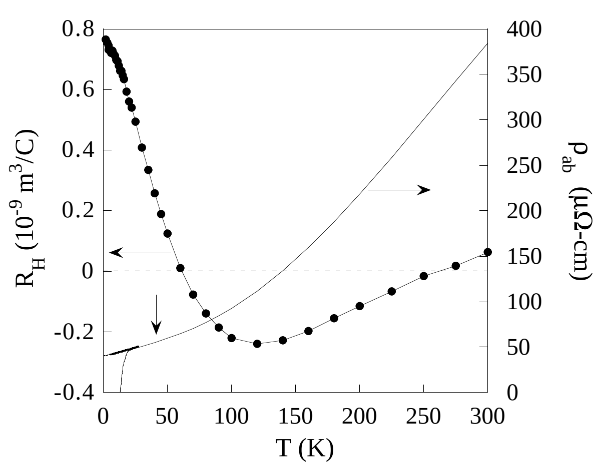

FIG. 4. Resistivity (at 0, 8.7, and 12 T) and Hall coefficient of the overdoped $x = 0.17$ film. The vertical arrow indicates the linear to quadratic crossover temperature (see text). The solid line for $R_H(T)$ is a guide to the eye.

coincides with the *positive* Hall coefficient at low temperature. Wang *et al.* [28] first showed that the positive low temperature Hall coefficient (R_H) of some $Nd_{1.85}Ce_{0.15}CuO_4$ crystals was consistent with two-carrier transport. This was later supported by other more detailed transport experiments [29,30]. The strong temperature dependence of R_H down to 2 K (despite the resistivity being constant) was a clear demonstration that carriers were still scattered inelastically at these temperatures. Wang *et al.* [28] showed that a fit to their $R_H(T)$ data gives a *linear* dependence of $1/\tau_{\rm hole}$ at low temperatures. In the case of our $x = 0.17$ thin films, it appears that the crossover, in this scenario, would correspond to the temperature where the conductivities are comparable for both negative (with $1/\tau_{\rm el} \propto T^2$) and positive (with $1/\tau_{\rm hole} \propto T$) carriers. Then the behavior of the positive carriers, dominating the resistivity at low temperatures, could be consistent with electron-electron scattering in a disordered 2D metal [31]. The same carriers could have a definite impact on the high-T resistivity, leading to observed deviations from the purely quadratic behavior. It is important to emphasize here the coincidence between the appearance of positive carriers, a linear component in the resistivity, and superconductivity. It is as if hole carriers are needed to get superconductivity in the copper-oxygen planes, a scenario suggested previously from the exhaustive transport data of Jiang *et al.* [29].

In summary, we observe a normal-state, $T = 0$, insulator-metal crossover as a function of Ce doping (x) in the electron-doped cuprates, similar to that found in the hole-doped ones. Measurements on overdoped samples ($x = 0.17$) give an unusual linear temperature dependence of the resistivity from 40 mK to 40 K and suggest electron-electron scattering in a disordered 2D metal of holelike carriers. The qualitative equivalence of the low temperature phase diagrams for the electron- and hole-doped cuprates suggests there is an electron-hole doping symmetry of the copper-oxygen planes. Our results underline the importance of understanding the electron-doped cuprates, and the need for a theory which can explain the similarities and the differences of the electron- and hole-doped cuprates.

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