Origin of the Anomalous Low Temperature Upturn in the Resistivity of the Electron-Doped Cuprate Superconductors

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The temperature, doping, and field dependences of the magnetoresistance (MR) in $Pr_{2-x}Ce_xCuO_{4-\delta}$ films are reported. We distinguish between orbital MR, found when the magnetic field is applied perpendicular to the *ab* planes, and the nearly isotropic spin MR. The latter, the major MR effect in the superconducting samples, appears in the region of the doping-temperature phase diagram where $d\rho/dT < 0$, or an upturn in the resistivity appears. We conclude that the upturn originates from spin scattering processes.

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Fermi-liquid theory generally describes the normal state of conventional superconductors. In the high-T_c cuprates, both the normal and the superconducting states depend on the carrier concentration in the CuO_2 planes (doping). In hole doped (*p*-doped) cuprates the overdoped region is believed to be metallic (Fermi-liquid-like), whereas in the underdoped region at low temperatures the resistivity increases with decreasing temperatures and may even be logarithmically diverging at $T = 0$ [1]. A similar behavior with decreasing doping is found in the electron-doped (*n*-doped) cuprates [2]. For optimally doped and underdoped samples when T is decreased from high temperature, the resistivity reaches a minimum at $T_{\min}$ and then begins to increase. $T_{\min}$ increases with decreasing doping. The origin of this anomalous upturn in resistivity has not been determined, and this is the main subject of this Letter.

The normal state of the *n*-doped cuprates is also characterized by negative magnetoresistance (*n*-MR) at low temperatures. Fournier *et al.* [3] interpreted the upturn in resistivity, as well as the *n*-MR, as a result of two-dimensional (2D) weak localization by disorder. In contrast, Seikitani *et al.* [4] suggested that the resistivity upturn and the *n*-MR are due to scattering off Cu^{2+} Kondo impurities induced by residual apical oxygen. In the *p*-doped cuprates, for example, negative MR was found in underdoped $La_{2-x}Sr_xCuO_4$ (LSCO) [5]. Other work was mainly focused on in-plain MR anisotropy in lightly doped LSCO [6], $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ [7], or nonsuperconducting $Pr_{2-x}Ce_xCuO_{4-d}$ (PCCO), $x = 0.15$ [8].

Recently, we found that the normal state Hall coefficient of PCCO at 350 mK exhibits a remarkable change at $x = 0.165 \pm 0.005$ [9]. This singular behavior was accompanied by significant changes in the temperature dependence of the resistivity below 20 K. These changes in the resistivity and Hall coefficient were suggested as strong evidence for a quantum critical point (QCP) at $x_c = 0.165 \pm 0.005$. We also found that below x_c the upturn in resistivity appears at low temperatures. The broad antiferromagnetic (AFM) region from $x = 0$ to just above $x = 0.15$ found in the phase diagram of the *n*-doped cuprates [10–12] suggests that the QCP found in Ref. [9] can be associated with the disappearance of the AFM phase as the doping is increased at $T = 0$.

In this Letter we report a new effect: an almost isotropic, spin related MR in PCCO, which vanishes suddenly above $x = 0.16$ at $T = 1.5$ K. While an orbital MR exists in the whole doping range and at a far different temperature scale than that of the resistivity upturn, the spin component exists only below $x = 0.16$ and has the same temperature scale as the upturn. We therefore conclude that the resistivity upturn is due to spin scattering. It may be related to AFM, which is found to persist well into the superconducting dome [11].

The samples are *c*-axis oriented PCCO thin films: $x = 0.11$, 0.12, 0.13, 0.14, 0.15, 0.16, 0.17, 0.18, and 0.19, whose preparation and characterization procedures were described elsewhere [9]. Measurements in the National High Magnetic Field Laboratory (NHMFL) were taken in a 32.4 T magnet and at temperatures ranging from 1.5 to 20 K. Other measurements were taken using a Quantum Design PPMS 14 T magnet. The field was aligned parallel to the *ab* planes ($H \parallel ab$) with an accuracy better than 0.25°. To exclude eddy current heating effects we ensured that the data were reproducible, symmetric for positive and negative magnetic fields, and independent of the sweeping rate. We measure the *ab*-plane resistivity with a standard four-probe technique.

In Fig. 1 we show the field dependence of the resistivity for two doping levels, $x = 0.15$ and $x = 0.16$, with field applied perpendicular to the *ab* planes ($H \perp ab$). Above a certain field, necessary to mute superconductivity, both samples exhibit *n*-MR at low temperatures. The amplitude of the *n*-MR decreases as the temperature increases and eventually vanishes around 8–10 K. The *n*-MR is found in the entire doping range studied (data not shown).

An immediate conclusion drawn from Fig. 1, and from the existence of *n*-MR for $H \perp ab$ for all x studied, is that there is no direct relation between the *n*-MR for $H \perp ab$

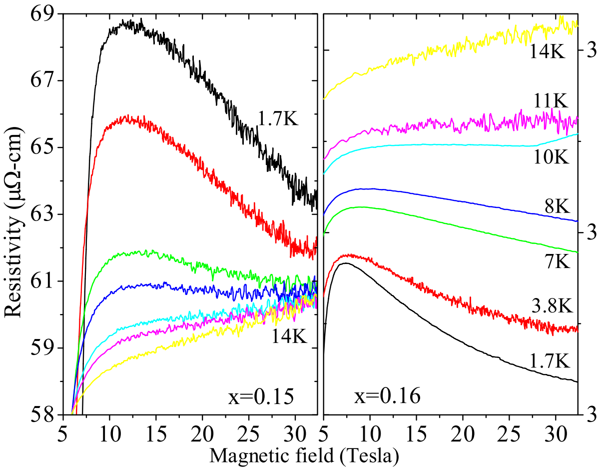

FIG. 1 (color online). The ab-plane resistivity of $\mathrm{Pr}_{2-x}\mathrm{Ce}_x\mathrm{CuO}_{4-\delta}$ films vs magnetic field applied perpendicular to the ab plane ($H \perp ab$).

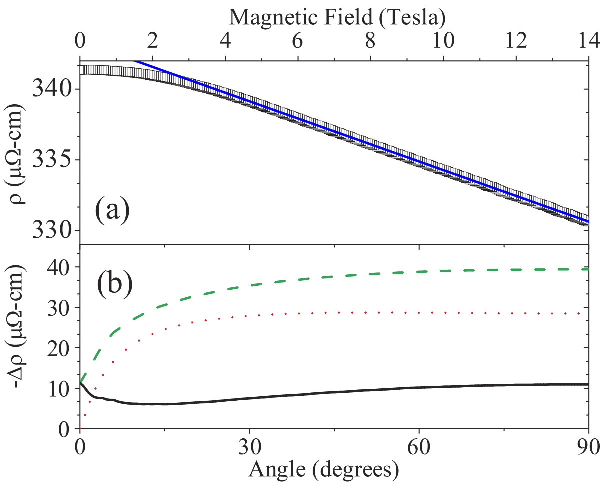

FIG. 2 (color online). The $x = 0.11$ sample at $T = 1.8$ K. (a) The resistivity vs field at $\theta = 0$ ($H \parallel ab$). Solid line is a linear fit to the high fields region. (b) Calculation of the angular dependence of a spin effect; see text for details.

and the upturn in resistivity. First, the upturn in resistivity appears only for $x \leq 0.16$ while the n-MR with $H \perp ab$ appears for $0.11 \leq x \leq 0.19$. Second, the upturn in resistivity and the onset of the n-MR with $H \perp ab$ occur at unrelated temperatures. For example, in $x = 0.15$ the $T_{\mathrm{min}} \approx 20$ K, while the n-MR with $H \perp ab$ vanishes around 8 K. For the $x = 0.16$ we find $T_{\mathrm{min}} < 1$ K, while the n-MR for $H \perp ab$ is observed up to 8 K. In this Letter, we concentrate on the negative longitudinal MR (LMR) due to the field component parallel to the ab planes. We show that the LMR is, in fact, a result of an isotropic effect, and it closely relates to the resistivity upturn: it vanishes at the same doping level and approximately the same temperatures at which the upturn disappears.

The resistivity versus field, $H \parallel ab$, is shown in Fig. 2(a) for the nonsuperconducting ($T_c < 0.35$ K) $x = 0.11$ film at 1.8 K. We now follow this LMR as a function of temperature and doping. In Fig. 3(a) we show the LMR as a function of field at various temperatures for the $x = 0.13$ sample. The LMR is negative at low temperatures and changes to positive above 80 K. In Fig. 3(b) we show the resistivity for $x = 0.13$ as a function of temperature. Upon fitting the resistivity to a form $\rho = \rho_0 + AT^{\beta}$ we find at high temperatures ($T > 120$ K) $\beta = 2$, and as the temperature is decreased β gradually decreases. The resistivity goes through a minimum at $T_{\mathrm{min}} \approx 60$ K, which is rather close to the temperature at which the LMR changes sign. However, it is difficult to say exactly where the upturn begins. To give a better estimate for this temperature we plot the effective resistivity exponent, $\beta = \frac{d\ln(\rho-\rho_0)}{d\ln T}$ as a function of temperature [circles, right hand scale in Fig. 3(b)]. ρ_0 is calculated from fitting the resistivity in the range $120 < T < 250$ K to the form $\rho = \rho_0 + AT^2$. Around 90 K β starts falling rapidly from its value at high temperatures, $\beta = 2$, to 0 at the minimum. This temperature is very close to the temperature at which LMR crosses over from positive to negative. Similar results were found in the $x = 0.11$ film. For higher dopings it is difficult to make such a measurement since the upturn begins close to ($x = 0.15$) or below T_c ($x = 16$), and a parallel field of 14 T is not sufficient to mute the affects of the superconductivity.

We have demonstrated that the upturn in resistivity and the LMR appear at the same characteristic temperature. We now show that they also have the same characteristic doping. Since the upper critical field for PCCO for $H \parallel ab$ is

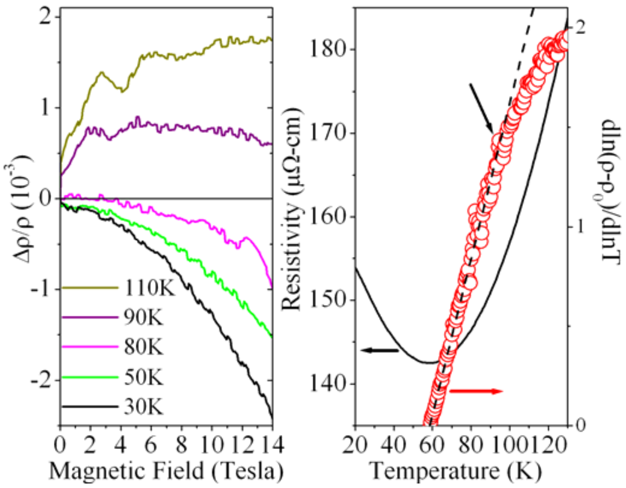

FIG. 3 (color online). (a) The temperature dependence of the LMR ($H \parallel ab$) for the $x = 0.13$ sample at various temperatures. Note that the MR turns positive above 80 K. (b) The resistivity as a function of temperature for the same sample (solid line, left hand scale). The effective exponent $d\ln(\rho - \rho_0)/d\ln T$ (circles, right hand scale) changes rapidly around 90 K, i.e., where the LMR changes sign. The dashed line is a guide to the eye.

greater than 32.4 T, it is impossible to directly measure the LMR and study its doping dependence at low temperatures. However, we can carry out another procedure that gives similar information. First, we measured the resistance as a function of $H \perp ab$ up to 32.4 T at 1.5 K. We then rotate the film in a field of 32.4 T with θ the angle between the field and the ab planes. $\theta = 0$ corresponds to the field applied parallel to the current (and the ab planes); $\theta = 90°$ is a field perpendicular to the film. If only $H_\perp$ affects the MR, then the resistance at a field H_0 applied perpendicular to the film should be the same as when the maximal available field of 32.4 T is applied at an angle $\theta_0 = \arcsin(H_0/32.4\ \mathrm{T})$. We therefore transform the field sweep into an effective angle, θ, using $\theta = \arcsin(H/32.4\ \mathrm{T})$.

The $H \perp ab$ sweep and the rotation in field measurements are compared in Fig. 4. Figures 4 and 5 are key figures in this Letter. In the high doping regime ($x = 0.16$) the results of the field sweep (green squares) and those from the rotation in the field (black circles) overlap. This means that there is *only* a perpendicular MR component, i.e., orbital MR. Similar results were obtained for the $x = 0.17$ and $x = 0.19$ samples (not shown). A completely different behavior is found in the $x = 0.15$–0.11 samples. There we can identify a strong parallel magnetic field effect, observed as the difference between the field sweep and the rotation in field. In fact, *the MR is almost isotropic in the* $x = 0.13$–0.15 *samples* (black circles). For the $x = 0.15$, $x = 0.14$, and $x = 0.13$ samples the resistivity changes by about 8%, 2.5%, and 2.7% in the $H \perp ab$ sweep while it changes by only 3%, 0.3%, and 0.6%, respectively, in the rotation. Surprisingly, this suggests that most of the n-MR is seen when $H \perp ab$ is coming from an isotropic spin effect, and it is *not* due to orbital or 2D effects such as weak localization [3] or to a field-tuned 2D superconductor-to-insulator transition [13].

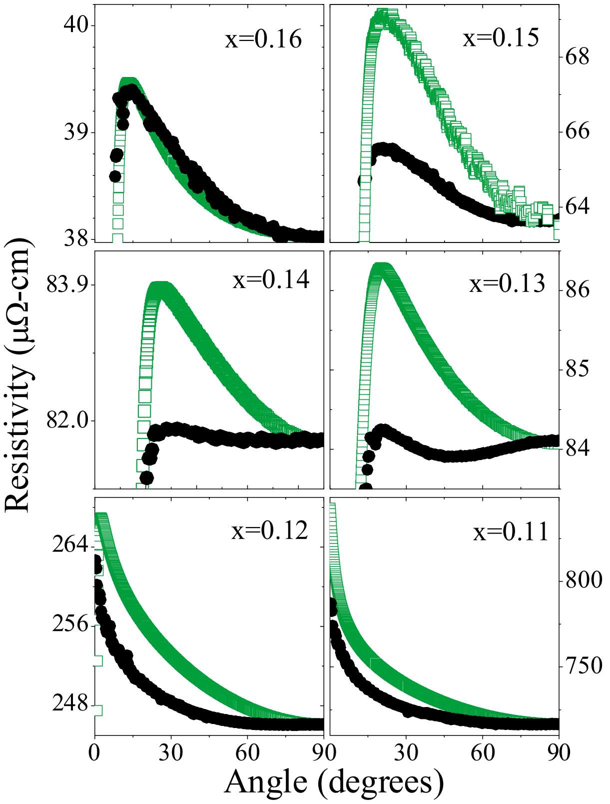

FIG. 4 (color online). Comparison between field sweeps $H \perp ab$ (green squares) and rotation in a magnetic field of 32.4 T (black circles) for various dopings at 1.5 K. The difference between the two measurements is due to the spin effect, which vanishes for $x \geq 0.16$ (see text).

In Fig. 5 we plot the difference between the resistance at 16.2 T ($H \perp ab$) [effective angle $\theta = \arcsin(16.2/32.4) = 30°$] and the resistance at 32.4 T applied at 30°, normalized with the resistance at 32.4 T ($H \perp ab$) versus doping (all at $T = 1.5$ K). This is a measurement of the isotropic spin effect of the additional 16.2 T. The spin effect vanishes dramatically at $x = 0.16$ at this temperature. This result is independent of the angle selected for the comparison between the field sweep and the rotation. This is consistent with the evidence for a quantum phase transition at $x = 0.165 \pm 0.005$ that we have previously reported [9]. In the $x = 0.16$ sample, the upturn can be seen only below 1 K; hence, the absence of a LMR component in this sample at 1.5 K is not surprising, and it is still consistent with a phase transition at $T = 0$ and $x_c = 0.165 \pm 0.005$. The sudden increase of $\Delta\rho(30°)/\rho(90°)$ at $x = 0.15$ is consistent with scattering off critical fluctuations near a quantum critical point.

Next, we further test our assumption of a coexistence of an anisotropic orbital effect and an isotropic spin effect. We use the $x = 0.11$ sample, where the absence of superconductivity allows us to measure the resistance as a function of parallel [Fig. 2(a)] and perpendicular fields from 0 to 14 T and the full rotation in a 14 T field to check this assumption (all measurements were taken at 1.8 K). In the following we use the notation $\bar{H} = (H_\perp, H_\parallel)$ where the

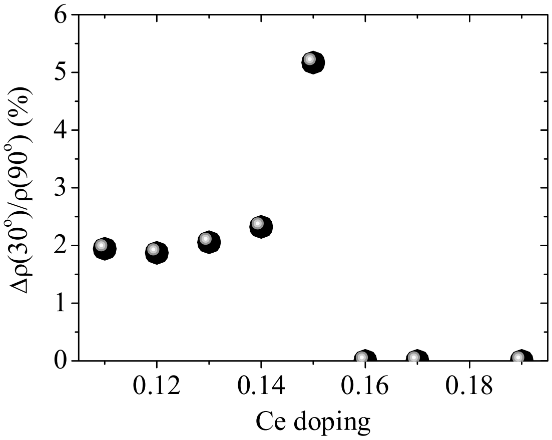

FIG. 5. The difference in resistivity between the field sweep and the rotation in field at 30° taken from Fig. 4 and normalized with the resistivity at 32.4 T ($H \perp ab$).

first (second) component is perpendicular (parallel) to the *ab* planes. When measuring the MR in perpendicular field configuration $\Delta\rho(H, 0)$, one picks up both the spin and the orbital effects, or $\Delta\rho(H, 0) = \Delta\rho_{\mathrm{orb}}(H, 0) + \Delta\rho_{\mathrm{spin}}(H, 0)$. However, in a measurement with a field applied parallel to the *ab* planes, one picks up only the spin effect $\Delta\rho(0, H) = \Delta\rho_{\mathrm{spin}}(0, H)$. Since the spin effect is isotropic, $\Delta\rho_{\mathrm{spin}}(0, H) = \Delta\rho_{\mathrm{spin}}(H, 0)$, one can calculate the orbital effect $\Delta\rho_{\mathrm{orb}}(H,0) = \Delta\rho(H,0) - \Delta\rho_{\mathrm{spin}}(H,0) = \Delta\rho(H,0) - \Delta\rho(0,H)$. From $\Delta\rho_{\mathrm{orb}}(H, 0)$, one can calculate (polar representation) $\Delta\rho_{\mathrm{orb}}(\theta)$ at 14 T. Since only the perpendicular component matters, at 14 T $\Delta\rho_{\mathrm{orb}}(\theta) = \Delta\rho_{\mathrm{orb}}(14\ \mathrm{T}\sin\theta, 0)$ [see red dotted line in Fig. 2(b)]. Now, at a certain field (14 T in our case) and any angle the spin MR is $\Delta\rho_{\mathrm{spin}}(\theta) = \Delta\rho(\theta) - \Delta\rho_{\mathrm{orb}}(\theta)$ [$\Delta\rho(\theta)$, again, picks up both the spin and the orbital effects]. The direct measurement $\Delta\rho(\theta)$ (dashed green line) and the resulting $\Delta\rho_{\mathrm{spin}}(\theta)$ at 14 T (black solid line) are plotted in Fig. 2(b) as a function of the angle. We note that $\Delta\rho_{\mathrm{spin}}$ is almost isotropic, consistent with a spin scattering mechanism. This is a verification of our assumption.

We note that Cieplak *et al.* [14] studied MR in lightly (hole) doped $\mathrm{La}_{2-x}\mathrm{Sr}_x\mathrm{CuO}_4$ films. They have found a spin related, *n*-MR effect, interpreted as a magnetic field suppression of spin disorder scattering. A more detailed doping study in the hole doped side is needed in order to determine if this effect is similar to the spin effect we report here.

We have shown here a correlation between the upturn and the isotropic, spin related MR, and, therefore, we suggest that the upturn is a result of a spin effect. A straightforward explanation could be a partial gapping of the Fermi surface due to AFM correlations as seen in angular resolved photoemission spectroscopy [15] and in optics [16]. However, the Nèel temperatures reported for $x = 0.10$ and $x = 0.13$ are 150 and 120 K, respectively. These temperatures are higher than the upturn-LMR temperatures that we find here. On the other hand, Woods *et al.* [17] irradiated an optimally doped $\mathrm{Nd}_{1.85}\mathrm{Ce}_{0.15}\mathrm{CuO}_4$ film and found an increase of T_{min} with increasing disorder. Therefore, we speculate that the upturn and the spin MR are related to magnetic impurity scattering which appears only in the AFM side of the phase diagram. More quantitative calculations are needed to confirm this suggestion.

In summary, doping, temperature, and field dependences of the *ab*-plane magnetoresistance were measured in $\mathrm{Pr}_{2-x}\mathrm{Ce}_x\mathrm{CuO}_{4-\delta}$ epitaxial films. An orbital magnetoresistance component exists throughout the whole doping range measured, $0.11 \leq x \leq 0.19$. Its temperature dependence is very different from that of the anomalous resistivity upturn. By contrast, a spin related magnetoresistance exists in the underdoped side. It vanishes dramatically at $x = 0.16$. This spin related MR is observed in the same doping and temperature ranges as the resistivity upturn. We therefore conclude that the anomalous upturn in resistivity is due to spin scattering.

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