# Spin-Flop Transition and the Anisotropic Magnetoresistance of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$: Unexpectedly Strong Spin-Charge Coupling in the Electron-Doped Cuprates

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(Received 20 October 2003; published 4 June 2004)

We use transport and neutron-scattering measurements to show that a magnetic-field-induced transition from noncollinear to collinear spin arrangement in adjacent $CuO_2$ planes of lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ ($x = 0.01$) crystals affects significantly both the in-plane and out-of-plane resistivity. In the high-field collinear state, the magnetoresistance (MR) does not saturate but exhibits an intriguing fourfold-symmetric angular dependence, oscillating from being positive at $\mathbf{B}\|[100]$ to being negative at $\mathbf{B}\|[110]$. The observed MR of more than 30% at low temperatures induced by a modest modification of the spin structure indicates an unexpectedly strong spin-charge coupling in electron-doped cuprates.

DOI: 10.1103/PhysRevLett.92.227003 PACS numbers: 74.25.Fy, 74.20.Mn, 74.72.Jt, 75.25.+z

High-$T_c$ superconductivity (SC) in cuprates emerges when the parent antiferromagnetic (AF) insulator is doped with charge carriers, which can be either holes or electrons. Despite this apparent symmetry with respect to doping, it still remains unclear whether the mechanism of SC in both cases is the same. It is generally believed that in the hole-doped cuprates, the SC pairing originates from an interplay between the doped holes and AF spin correlations. Indeed, many observations, including a fast suppression of the Néel order by doped holes [1], which results in the "spin-glass" state [1–3] and a strong tendency to form spin-charge textures or "stripes" [4], point to a very strong coupling between the charge and spin degrees of freedom.

The behavior of doped *electrons* looks, however, much different. Electron doping suppresses the AF order at virtually the same slow rate as the substitution of magnetic $Cu^{2+}$ ions with nonmagnetic $Zn^{2+}$ [5,6] and does not induce any incommensurability in the spin correlations [7]. This has been taken as evidence that the electrons merely dilute the spin system [1,5,6]. Apparently, if the charge transport and spin correlations are actually decoupled in the electron-doped cuprates, the SC pairing should have a nonmagnetic origin as well. A recent discovery of the magnetic-field induced AF order in superconducting $Nd_{2-x}Ce_xCuO_4$ [8,9] has shown, however, that antiferromagnetism and superconductivity may be closely related in these compounds.

To probe the spin-charge coupling, one can determine how the charge transport responds to such relatively weak changes in the spin structure as spin-flop or metamagnetic transitions. In hole-doped $La_{2-x}Sr_xCuO_4$, for instance, the conductivity changes by up to several times [10,11]. In this Letter, we use neutron-scattering and magnetoresistance (MR) measurements to study the effect of magnetic field on the spin structure and anisotropic conductivity of lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ (PLCCO) single crystals. We find that both the in-plane and out-of-plane resistivity ($\rho_{ab}$ and $\rho_c$) are surprisingly sensitive to spin reorientation, with $\Delta\rho_{ab}/\rho_{ab}$ exceeding 30% at low temperatures—the same scale as in hole-doped $La_{2-x}Sr_xCuO_4$ [11]. This result indicates that in electron-doped cuprates the charge transport exhibits a similar degree of coupling to magnetism as in the hole-doped ones, and therefore the superconductivity in both systems may have a universal origin.

High-quality PLCCO single crystals with $x = 0.01$ (mosaicity $<1°$) were grown by the traveling-solvent floating-zone technique and annealed at $\approx 860\,°C$ in pure argon to remove excess oxygen. The partial substitution of Pr with La was used to stabilize the crystal growth without introducing significant lattice distortions [12]. Neutron-scattering measurements were performed on the BT-2 and SPINS triple-axis spectrometers at the NIST Center for Neutron Research. We label wave vectors $\mathbf{Q} = (q_x, q_y, q_z)$ in $Å^{-1}$ as $(H, K, L) = (q_x a/2\pi, q_y a/2\pi, q_z c/2\pi)$ in the reciprocal lattice units (r.l.u.) suitable for the tetragonal unit cell of PLCCO (space group $I4/mmm$, $a = 3.964$ and $c = 12.28$ Å are in-plane and out-of-plane lattice parameters, respectively). In this notation, [100]/[010] and [110]/[$\bar{1}$10] are along the Cu-O-Cu bond direction and the diagonal Cu-Cu direction, respectively. The experimental details are described in Refs. [8,9].

Resistivity measurements were carried out by the ac four-probe method on the *same* crystal used for neutron measurements. It was cut and polished into suitable shapes: $3.1 \times 1 \times 0.45\ mm^3$ for $\rho_{ab}$ and $\approx 1 \times 1 \times 1\ mm^3$ for $\rho_c$. The MR was measured by sweeping the magnetic field between $\pm 14$ T at fixed temperatures

  

stabilized by a capacitance sensor with an accuracy of ~1 mK.

The peculiar spin structure of $Pr_2CuO_4$ (PCO) is interesting in its own right. While a strong intraplane exchange drives the AF spin ordering within $CuO_2$ planes, all the isotropic exchange interactions *between* the planes are perfectly canceled out due to the body-centered tetragonal crystal symmetry. The three-dimensional ordering [Fig. 1(a)] that sets in below the Néel temperature $T_N = 250$–$285$ K [13–15] is governed by weak pseudodipolar (PD) interactions, which favor a noncollinear orientation of spins in adjacent planes (alternating along the [100] and [0$\bar{1}$0] directions) [13–16]. A unique feature of the interplane PD interaction is that its energy does not change if the spin sublattices of adjacent $CuO_2$ planes rotate in *opposite* directions [14–16]. Such a continuous spin rotation can be induced by a magnetic field parallel to Cu-Cu direction, which easily converts the noncollinear structure of Fig. 1(a) into a collinear one with spins along the [110] direction [Fig. 1(b)]. Note that, while these diagonal directions are hard spin axes in the noncollinear phase, they become the easy axes in the collinear one. A perfectly aligned field **B**||[010] causes a first-order transition directly to the spin-flop phase [Fig. 1(d)], while at intermediate field directions the magnetic field first induces a transition into the collinear phase [Fig. 1(c)] and then smoothly rotates the spins to align them perpendicular to the field [15].

The neutron diffraction measurements at zero field on the $(1/2, 1/2, L)$ magnetic Bragg peaks ($L = 0, 1, 2, 3, 4$) show that in our PLCCO ($x = 0.01$) the $Cu^{2+}$ spins order into the same noncollinear structure as in pure PCO, albeit at a somewhat lower $T_N \approx 229$ K (Fig. 2). The reduced $T_N$ is probably due to a partial substitution of $Pr^{3+}$ with nonmagnetic $La^{3+}$, as well as to doped electrons. Similar to PCO [13], the $Pr^{3+}$ ions in PLCCO can be polarized by the ordered $Cu^{2+}$ moment. Upon cooling below 100–150 K, the exchange field of the $Cu^{2+}$ spins induces a small (up to ~0.1 $\mu_B$) ordered moment on the $Pr^{3+}$ ions (Fig. 2).

Figure 3 shows the effect of a **B**||[$\bar{1}$10] field on the $(1/2, 1/2, 1)$ and $(1/2, 1/2, 2)$ magnetic peaks at various temperatures. Upon increasing the magnetic field, the peak intensity changes, indicating a continuous noncollinear to collinear phase transition. Indeed, for the collinear spin arrangement [Fig. 1(b)], the magnetic intensity vanishes at $(1/2, 1/2, L)$, with $L = 1, 3, 5$. As can be seen in Figs. 3(c) and 3(d), the critical field for the noncollinear to collinear ("spin-flop") transition, $B_c$, increases from less than 0.5 T at 150 K to ~2 T at 5 K. In comparison, the first-order spin-flop transition for **B**||[010] was reported to take place at several times larger fields [15], and a $c$ axis aligned field does not change the noncollinear spin structure [9].

The transport properties of lightly electron-doped PLCCO differ from those of its hole-doped analog LSCO or $YBa_2Cu_3O_{6+x}$ (YBCO). In contrast to hole-doped cuprates [11,17,18], the doping of 1% of electrons into the $CuO_2$ planes appears to be insufficient to induce metallic in-plane conduction in PLCCO, and both $\rho_{ab}$ and $\rho_c$ grow upon cooling below room temperature [Fig. 4(a)]. It is worth noting also that lightly doped PLCCO turns out to be one of the most anisotropic cuprates with $\rho_c/\rho_{ab} \sim 8000$ at room temperature—an order of magnitude larger than in LSCO and YBCO [11,18].

In further contrast to hole-doped cuprates [11,18], no anomaly is detected at the Néel transition in PLCCO either in the in-plane or out-of-plane resistivity. At a first glance, this supports the view that the charge motion in

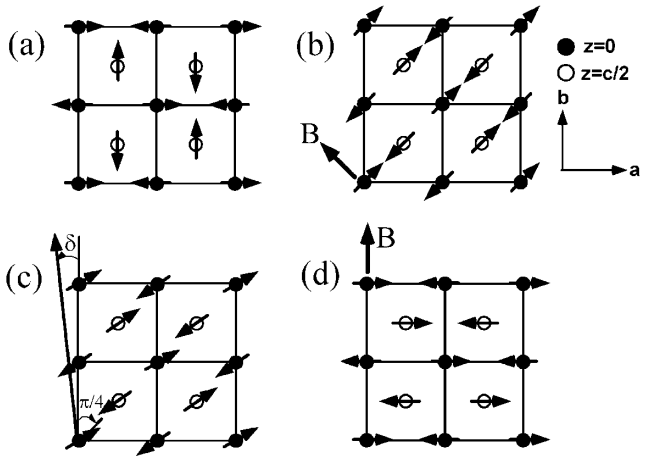


FIG. 1. Field-induced transition from noncollinear to collinear spin arrangement in $Pr_2CuO_4$. (a) Zero-filed noncollinear spin structure; only Cu spins are shown. (b),(c) Collinear spin-flop states induced by (b) a magnetic field applied along the Cu-Cu direction. (c) A magnetic field tilted from [010]. (d) **B**||[010].

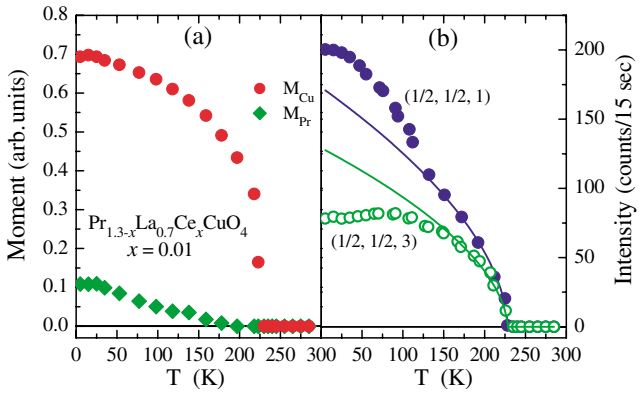


FIG. 2 (color online). (a) Temperature dependence of the $Cu^{2+}$ and $Pr^{3+}$ moments in PLCCO ($x = 0.01$). (b) Integrated intensity of the $(\frac{1}{2}, \frac{1}{2}, 1)$ and $(\frac{1}{2}, \frac{1}{2}, 3)$ magnetic peaks. The ordered moments are estimated by normalizing the magnetic intensity to the weak $(1, 1, 0)$ nuclear Bragg peak without considering the absorption and extinction effects [9]. The solid lines are power-law fits describing the contribution of Cu spins [13].

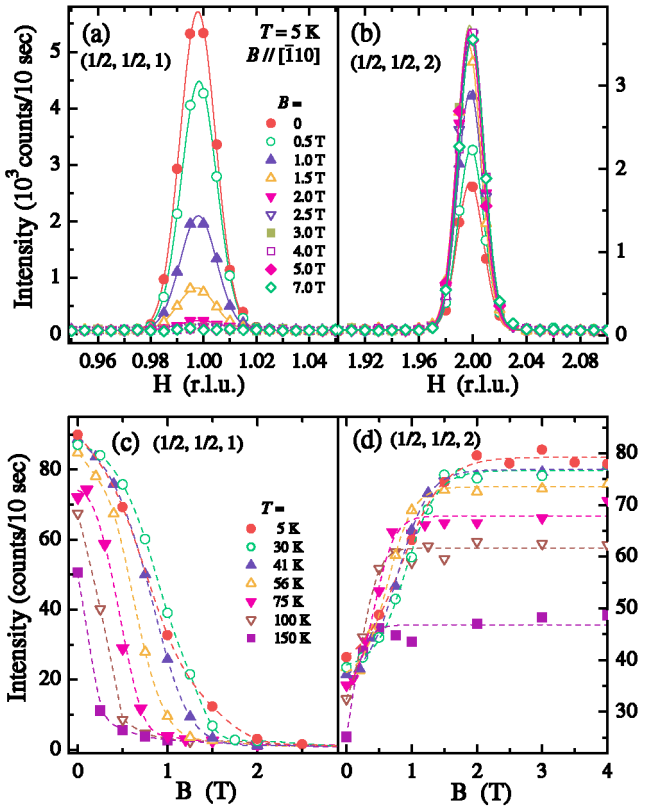


FIG. 3 (color online). (a),(b) Effect of the $\mathbf{B}\|[\bar{1}10]$ field on $(\frac{1}{2}, \frac{1}{2}, 1)$ and $(\frac{1}{2}, \frac{1}{2}, 2)$ magnetic peaks at 5 K. (c),(d) Field dependence of the integrated intensity at various temperatures. We note that the critical field for spin-flop transition in PLCCO is lower than that of PCO [13].

electron-doped PLCCO is virtually decoupled from spin correlations, and one therefore would expect the conductivity to ignore the spin reorientation sketched in Fig. 1. Surprisingly, the experiment shows that this is not the case, and instead of being field-independent, both $\rho_{ab}$ and $\rho_c$ exhibit a considerable increase upon transition into the collinear state [Figs. 4(b) and 4(c)]. We have confirmed that this MR is of the spin origin and contains no orbital terms, since no difference was observed in $\Delta\rho_{ab}/\rho_{ab}$ for fields applied parallel or perpendicular to the current. Moreover, $\Delta\rho_{ab}/\rho_{ab}$ and $\Delta\rho_c/\rho_c$ demonstrate a remarkable similarity both in magnitude and in field dependence, in spite of the huge resistivity anisotropy. Finally, no MR anomaly is observed when a $c$ axis aligned field is applied, consistent with the absence of a spin-flop transition for such field orientation [9].

The MR behavior in Fig. 4 is clearly reminiscent of that in LSCO [11], though there are two important differences. First is the sign of the anomalous MR, which is always positive in PLCCO but negative in LSCO. Second, the MR features in LSCO and YBCO become discernible as soon as the AF order is established, but in PLCCO they appear at temperatures much lower than $T_N$ (at $T <$ 70–100 K), and quickly gain strength upon decreasing temperature (Fig. 4). The latter indicates that some other factors, such as magnetic moments of $Pr^{3+}$ or a structural instability [19], that come into play at low temperature may be relevant to the observed MR.

A comparison of the neutron and resistivity data reveals one more interesting feature; namely, the transitions observed by these two probes do not match each other [inset of Fig. 5(a)]. One can see that the charge transport ignores the initial spin rotation, and the steepest resistivity variation is observed at $B_c$, where the collinear structure is established. Although $B_c$ changes substantially with temperature [Fig. 5(a)], the apparent shift in the transitions holds consistently, with the peak in $d\rho/dB$ roughly coinciding with the end of the transition observed by neutron scattering.

As the magnetic field deviates from the Cu-Cu direction [Fig. 1(c)], the spin-flop transition shifts towards higher fields, ultimately reaching $B_c \sim 12$ T for $B\|[010]$; the MR behavior for these two field orientations is compared in Figs. 5(b) and 5(c) [20]. It becomes immediately clear from these figures that the steplike increase of the resistivity upon the transition to the

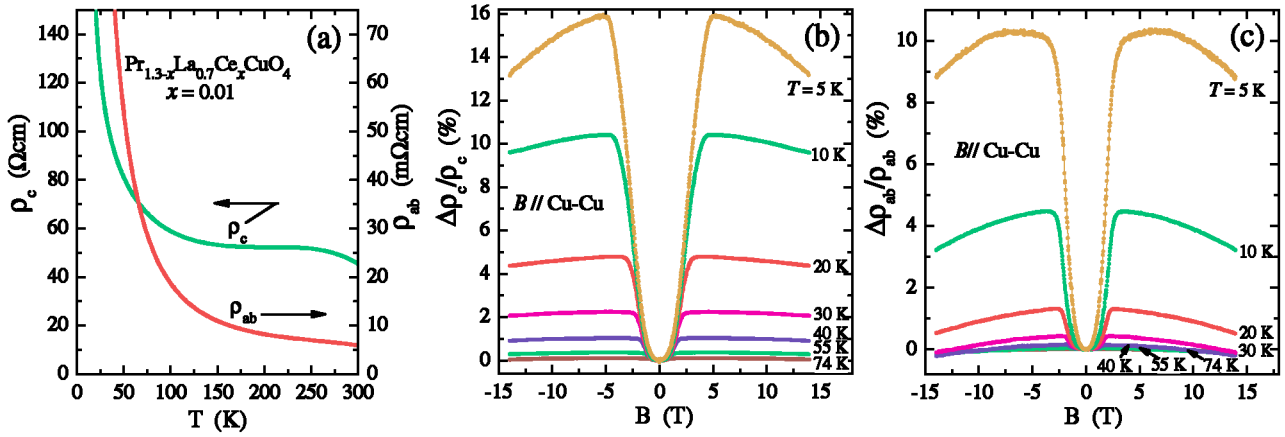


FIG. 4 (color online). (a) In-plane and out-of-plane resistivity of PLCCO ($x = 0.01$) single crystals. The MR in $\rho_c$ (b) and $\rho_{ab}$ (c) measured for the in-plane magnetic field $\mathbf{B}\|[\bar{1}10]$.

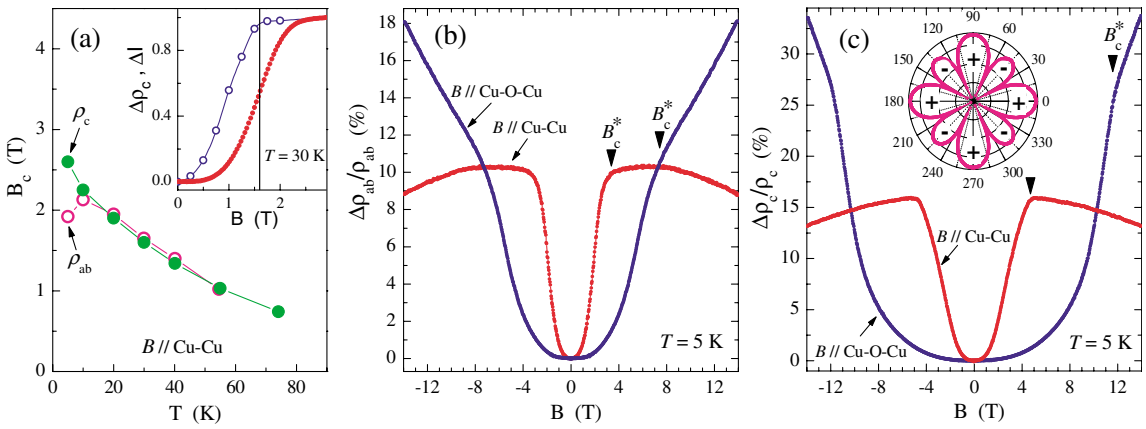


FIG. 5 (color online). (a) The critical field $B_c$ determined from peaks in $d\rho_{ab}/dB$ and $d\rho_c/dB$ for $\mathbf{B}\|[\bar{1}10]$. In the inset of (a), the normalized field dependence of $\rho_c$ (●) is compared with that of the $(\frac{1}{2}, \frac{1}{2}, 1)$-peak intensity (○). $\Delta\rho_{ab}/\rho_{ab}$ (b) and $\Delta\rho_c/\rho_c$ (c) for two directions of the in-plane magnetic field. The angular dependence of the high-field MR is sketched in the inset of (c).

collinear state does not make a complete story. Regardless of the field direction within the *ab* plane, the resistivity exhibits roughly the same increase at the spin-flop transition, but then (at $B > B_c^*$) it keeps changing without any sign of saturation [Figs. 5(b) and 5(c)]. Even more surprising is that this high-field MR changes its sign depending on the field direction, as is schematically drawn in the inset of Fig. 5(c). One can conceive a spin structure upon rotating the high magnetic field within the *ab* plane in the following way: the spins always keep the collinear arrangement and rotate as a whole, being almost perpendicular to the magnetic field (Fig. 1). Our data show that the resistivity goes down as the spin direction approaches one of the two equivalent spin easy axes (Cu-Cu directions) and increases at the spin hard axes (Cu-O-Cu directions) [inset of Fig. 5(c)]. Note that the resistivity changes are rather large, $\Delta\rho_{ab}/\rho_{ab}$ reaches $\approx 18\%$ at $T = 5$ K and exceeds 32% at 2.5 K, indicating that the magnetic field $\mathbf{B}\|[100]$ can effectively localize the doped electrons.

Apparently, the fascinating MR oscillations in Fig. 5 cannot originate from simple "spin-valve" effects, since at high fields the spin structure always stays collinear, and all that changes is the relative orientation of spins with respect to the crystal axes. The MR may be related to 2D spin fluctuations that were found to survive far above $B_c$, as manifested in the diffuse neutron scattering [21], or to some unusual coupling of the charge transport with low-energy spin dynamics. Though the exact mechanism of the revealed MR features still remains to be understood, what is certain is that the charge carriers in electron-doped cuprates appear to have a remarkably strong coupling with the spin order, which should play an important role in determining their physical properties.

Upon preparing this Letter, we became aware of similar MR features observed for $Pr_{1.85}Ce_{0.15}CuO_4$ [22], which gives evidence that the strong spin-charge coupling survives up to much higher electron-doping levels, which are relevant for the superconducting state.

We thank K. Segawa and Shiliang Li for technical assistance. This work was in part supported by the U.S. NSF Grant No. DMR-0139882 and DOE under Contract No. DE-AC-00OR22725 with UT/Battelle, LLC.

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