High-$T_c$ superconductivity (SC) in cuprates emerges as the parent antiferromagnetic (AF) insulator is doped with charge carriers, 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 from 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 $< 1^\circ$) were grown by the traveling-solvent floating-zone technique and annealed at $\approx 860^\circ$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 wavevectors $Q = (q_x, q_y, q_z)$ in $\mathbf{A}^{-1}$ 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 $[110]/[\overline{1}10]$ 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.
field between $\pm 14$ T at fixed temperatures stabilized by a capacitance sensor with an accuracy of $\sim 1$ mK.

The peculiar spin structure of Pr$_2$CuO$_4$ (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, 14, 15] is governed by weak pseudodipolar (PD) interactions, which favor a noncollinear orientation of spins in adjacent planes (alternating along the [100] and [010] directions) [12, 14, 15, 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, 15, 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 non-collinear phase, they become the easy axes in the collinear one. A perfectly aligned field $B \parallel [110]$ 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 [16].

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 non-collinear 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 non-magnetic 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 $\sim 0.1 \mu_B$) ordered moment
on the Pr$^{3+}$ ions (Fig. 2).

Figure 3 shows the effect of a $\mathbf{B} \parallel [\overline{1}0\overline{1}]$ 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 non-collinear to collinear phase transition. Indeed, for the non-collinear to collinear (“spin-flop”) transition, $B_c$, increases from less than 0.5 T at 150 K to $\sim 2$ T at 5 K. In comparison, the first-order spin-flop transition for $\mathbf{B} \parallel [010]$ was reported to take place at several time larger fields $\[16\]$ and a c-axis aligned field does not change the noncollinear spin structure $\[17\]$.

The transport properties of lightly electron-doped PLCCO differ from those of its hole-doped analog LSCO or YBa$_2$Cu$_3$O$_{6+x+y}$ (YBCO). In contrast to hole-doped cuprates $\[11\] \[18\] \[19\] \[20\]$, the doping of 1% of electrons into the CuO$_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 $\[11\] \[18\] \[19\] \[20\]$. In further contrast to hole-doped cuprates $\[11\] \[18\] \[19\] \[20\]$, 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 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 $\[20\]$.

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 that $T_N$ (at $T < 70 \sim 100$ 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 to 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, reaching ultimately $B_c \sim 12$ T for $\mathbf{B} \parallel [010]$; 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 step-like increase of the resistivity upon the transition to the collinear state does
FIG. 5: (a) The critical field $B_c$ determined from peaks in $d\rho_{ab}/dB$ and $d\rho_{c}/dB$ for $B \parallel [\bar{1}0]$. In the inset to (a), the normalized field dependence of $\rho_c$ ($\bullet$) is compared with that of the $(\bar{2}, \bar{2}, 1)$-peak intensity ($\circ$), $\Delta\rho_{ab}/\rho_{ab}$ (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 to (c).

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 to 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 to 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 $B \parallel [100]$ 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 paper, 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, that are relevant for the superconducting state.

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

* Also at Department of Physics, Tokyo University of Science, Shinjuku-ku, Tokyo 162-8601, Japan.
† Electronic address: daip@ornl.gov
‡ Electronic address: ando@criepi.denken.or.jp
[1] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. En-doh, Rev. Mod. Phys. 70, 897 (1998).
[2] Ch. Niedermayer et al., Phys. Rev. Lett. 80, 3843 (1998).
[3] A. N. Lavrov et al., Phys. Rev. Lett. 87, 017007 (2001).
[4] J. M. Tranquada et al., Nature 375, 561 (1995).
[5] B. Keimer et al., Phys. Rev. B 45, 7430 (1992).
[6] G. M. Luke et al., Phys. Rev. B 42, 7981 (1990).
[7] K. Yamada et al., Phys. Rev. Lett. 90, 137004 (2003).
[8] H. J. Kang et al., Nature 423, 522 (2003).
[9] M. Matsuura et al., Phys. Rev. B 68, 144503 (2003).
[10] T. Thio et al., Phys. Rev. B 38, 905 (1988).
[11] Y. Ando et al., Phys. Rev. Lett. 90, 247003 (2003).
[12] M. Fujita et al., Phys. Rev. B 67, 014514 (2003).
[13] I. W. Sumarlin et al., Phys. Rev. B 51, 5824 (1995).
[14] D. Petitgrand et al., Phys. Rev. B 59, 1079 (1999).
[15] R. Sachidanandam et al., Phys. Rev. B 56, 260 (1997).
[16] V. P. Plakhty et al., Europhys. Lett. 61, 534 (2003).
[17] Y. Ando et al., Phys. Rev. Lett. 87, 017001 (2001).
[18] A. N. Lavrov et al., Phys. Rev. Lett. 83 (1999) 1419.
[19] V. P. Plakhty et al., Solid State Commun. 103, 683 (1997).
[20] For the case of $B || [010]$, the transition field is extremely sensitive to the crystal alignment; just a few degrees tilting of the $\rho_{ab}$ sample in Fig. 5(b) notably shifted the transition to low fields, as compared with the better aligned $\rho_{c}$ sample in Fig. 5(c).
[21] D. Petitgrand et al., App. Phys. A. 74, S853 (2002).
[22] P. Fournier et al., cond-mat/0309144.