Fourfold oscillations and anomalous magnetic irreversibility of magnetoresistance in the non-metallic regime of Pr$_{1.85}$Ce$_{0.15}$CuO$_4$

P. Fournier, M.-E. Gosselin, S. Savard, J. Renaud, I. Hetel, P. Richard and G. Riou

Département de Physique, Université de Sherbrooke,
Sherbrooke, Québec, CANADA, J1K 2R1

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Abstract

Using magnetoresistance measurements as a function of applied magnetic field and its direction of application, we present sharp angular-dependent magnetoresistance oscillations for the electron-doped cuprates in their low-temperature non-metallic regime. The presence of irreversibility in the magnetoresistance measurements and the related strong anisotropy of the field dependence for different in-plane magnetic field orientations indicate that magnetic domains play an important role for the determination of electronic properties. These domains are likely related to the stripe phase reported previously in hole-doped cuprates.

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In the low doping regime of high temperature superconductors (HTSC), resistivity crosses over most often from a high temperature metallic-like state to a low temperature non-metallic behavior\[1\]. Interestingly, the crossover temperature observed by in-plane resistivity does not correspond to the Néel temperature ($T_N$), the onset of long range antiferromagnetic order. In fact, there is no hint of this transition in $\rho_{xx}(T)$ as metallic-like behavior persists sometimes below $T_N$ \[2, 3\]. Ando and coworkers argued that this independence of $\rho_{xx}(T)$ from the underlying magnetic order is a sign of phase separation, probably with conducting stripes separated by antiferromagnetic domains. Such self-organized structures of the charge carriers was shown to lead to intriguing behaviors for several physical properties\[4, 5, 6, 7\]. An anisotropic twofold response to magnetic field applied along the copper-oxygen ($\text{CuO}_2$) planes was reported for instance using in-plane magnetoresistance (MR) in the non-metallic regime of YBa$_2$Cu$_3$O$_{6.33}$ (YBCO)\[4\] and in lightly doped La$_{1.99}$Sr$_{0.01}$CuO$_4$ (LSCO)\[5\].

To avoid the possible contributions of orthorhombic distortions or phases (as in YBCO and LSCO), we have chosen the tetragonal electron-doped cuprates\[8\] as good candidates to make a definitive test of anisotropic transport properties. We focus on the results obtained for magnetic fields always applied parallel to the CuO$_2$ planes. In Figure 1, we present an example of raw in-plane and out-of-plane resistance data as a function of angle with respect to the in-plane crystal a-axis. These data were measured deep into the non-metallic regime of non-superconducting Pr$_{1.85}$Ce$_{0.15}$CuO$_4$ (PCCO). We observe clear fourfold and symmetrical oscillations of the resistance with sharp maxima [minima] for $R_{xx} [R_c]$ for fields applied along the in-plane crystal axis (i.e. along the CuO bonds of the CuO$_2$ planes) and broad minima [maxima] for field applied along the diagonals. This effect can be emphasized using the polar plot of Fig. 1(c).

In this Letter, we present this magnetoresistance anisotropy, for current along the CuO$_2$ planes and along the c-axis (thickness) of non-superconducting electron-doped single crystals. We show for the first time that sharp fourfold oscillations persist over the whole non-metallic regime for both in-plane ($\rho_{xx}$) and out-of-plane ($\rho_c$) resistivities. More importantly, the field dependence of $\rho_c$ presents irreversibilities which cannot be explained by conventional band theory. We ascribe the new transport signatures to the presence of magnetic domains. Their origin is probably related to the presence of stripe domains (order) in the cuprates.

The single crystals of Re$_{2-x}$Ce$_x$CuO$_4$ (Re = Pr, Nd, Sm and Eu) used for this study were grown by the directional flux growth method in alumina and high purity magnesia.
FIG. 1: Raw resistance data as a function of angle for non-superconducting Pr$_{1.85}$Ce$_{0.15}$CuO$_4$:
(a) in-plane resistance $R_{xx}$, (b) out-of-plane resistance $R_c$ and (c) Polar plot of the same data. 0°
corresponds to the field applied along the Cu-O bonds. The inserts in (a) and (b) show the contact
configurations used for the measurements.

Silver epoxy contacts were applied directly onto the as-grown crystals (typical in-plane
size : 2mm x 1 mm, and 30 µm thickness along the c-axis) in two different configurations
insuring uniform current density for the measurement of the in-plane ($\rho_{xx}$) and c-axis ($\rho_c$)
resistivity as shown in the inserts of Figs. 1(a) and (b). The samples were then mounted on
sapphire supports and attached onto a specially designed Physical Property Measurement
System (PPMS) rotator chip, such that the applied magnetic field can be rotated over a
FIG. 2: (a) In-plane resistivity and c-axis resistivity as a function of temperature for non-superconducting Pr$_{1.85}$Ce$_{0.15}$CuO$_{4+\delta}$ crystals. (b) in-plane magnetoresistance as a function of field for $T = 5K$ and its anisotropy for field applied along the c-axis ($\perp$) and along the CuO$_2$ planes($||$).

The direction of the in-plane crystal axis (a-axis direction relative to the edge of the rectangular crystals) were determined using Laue x-ray diffraction and by Raman scattering. Several tests were performed to rule out contact configuration, thermometry and misorientation problems.

In Figure 2(a), we present the typical temperature dependence of in-plane and c-axis resistivity components for non-superconducting Pr$_{1.85}$Ce$_{0.15}$CuO$_4$ single crystals. Both components show a non-metallic regime at low temperatures where the MR oscillations are observed. The in-plane resistivity $\rho_{xx}$ is metallic-like at room temperature while it shows an upturn starting at $T_{\text{min,xx}} \approx 125K$ which is very sensitive to the oxygen content. Below $T_{\text{min,xx}}$, $\rho_{xx}$ approaches a $\ln(T)$ behavior (not shown), similar to that reported for non-superconducting NCCO and PCCO[11]. This temperature behavior and the strong anisotropy illustrated in Fig. 2(b) have been interpreted as a signature of two-dimensional weak localization (2DWL) by disorder[12]. For the c-axis resistivity $\rho_c$, a non-metallic trend is observed at room temperature for several crystals, followed by a maximum
$T \approx 200K$ [13], then by a metallic-like regime. It is finally followed by a non-metallic regime, also approaching $\ln(T)$, below $T_{\text{min,c}} \approx 70K$.

The field dependence of the in-plane magnetoresistance oscillations at 10K are presented in Fig. 3(a). The MR oscillations evolve steadily from broad features at 2T to sharp maxima for 4T whenever the field is applied along the Cu-O bonds. For fields approaching 9T, new maxima develop for fields applied approximately along the diagonal directions. The relative proportion of both oscillations varies also steadily with temperature as evidenced by the same 45° features barely appearing at 5T and 2K in Fig. 3(b) (see arrows). The amplitude of these oscillations in $\rho_{xx}$ represents only a small fraction ($\Delta \rho_{\text{osc}}/\rho \sim 0.05\%$) of the total negative MR [$\Delta \rho_{\text{tot}}/\rho \sim 1\%$ : see Fig. 2(b)]. Thus, unlike YBCO and LSCO, $\rho_{xx}$ oscillations are very weak and have almost perfect fourfold symmetry.

In Fig. 3(b), we present the evolution of these $\rho_{xx}$ oscillations (at 5T) with temperature. A similar effect is obtained if one decreases the temperature or increases the applied field. We observe that the weak oscillations disappear quickly with increasing temperature, vanishing close to $T_{\text{min,xx}}$. This seems to imply that this anisotropic behavior can only be observed in
FIG. 4: Out-of-plane magnetoresistance as a function of angle: (a) for several fields at 5K; (b) and (c) for several temperatures at 5T. The data has been moved vertically for clarity.

the non-metallic regime of these materials (within our sensitivity).

In Fig. 4(a), we present the angular dependence of $\rho_c$ in several magnetic fields at 5K. We observe very sharp minima in the c-axis resistivity, developing particularly for intermediate fields (3 to 6T). These sharp cusps appear for magnetic fields applied along the Cu-O bonds. As the applied magnetic field is further increased, these anomalies are gradually replaced by oscillations of smaller amplitude. In Figs. 4(b) and (c), we illustrate the strong temperature dependence of these features as they seem once again to vanish as the sample reaches temperatures close to the crossover to the metallic-like state. As will be shown below, the magnitude of the oscillations observed for $\rho_c$ are comparable to the total positive MR, in sharp contrast with those observed with $\rho_{xx}$. Therefore, they are less sensitive to mis-orientation of the crystals and thus easily observed.

In Figure 5 (a), we present the c-axis magnetoresistance at $T = 5K$ as a function of in-plane magnetic field applied along three different directions (0, 15 and 45°). Contrary to the in-plane resistivity, the c-axis magnetoresistance is positive and presents an unusual field dependence. For the field applied along the a-axis (at 0°), the resistivity remains remarkably
flat at low fields until a threshold field is reached. At this point, the resistance varies sharply, as if the system was crossing a transition. Beyond this threshold, the magnetoresistance resumes a high field $-B^2$ behavior. As soon as the field direction deviates from the a-axis [for 15 and 45° in Fig. 5(a)], the magnetoresistance presents a very sharp positive increase at low fields, quickly reaching the saturation $-B^2$ regime. The large magnitude of the oscillations in $\rho_c$ at about 4 - 5T in Fig. 4 (a) can easily be explained by the strong variations in resistance at 5T for different field orientations underlined by the dashed line in Fig. 5(a). We should emphasize here that $\rho_{xx}$ displays its sharpest peaks in the same range of applied field.

Interestingly, upon decreasing the applied magnetic field, the resistance for $\theta = 0^\circ$ shows a similar transition-like pattern, but the resistance presents a clear sign of irreversibility observed in Fig. 5(b). The arising of magnetic hysteresis is a clear demonstration of the influence of magnetic history, i.e. the presence of magnetic domains. These domains (and their domain walls) are strongly affected by the direction and strength of the in-plane rotat-
ing field, and in turn they affect the conductivity. We can interpret the data in Fig. 5(a) as evidence of easy magnetization axis along the diagonal directions, while the Cu-O bond directions (a and ”b” axis) correspond to hard axis. Since the c-axis resistivity is a measure of interplane tunneling between CuO$_2$ planes, the alignment of magnetic domains at high fields is *detrimental* to interplane tunneling causing an increase of c-axis resistivity. This effect is present up to temperatures approaching $T_{\text{min,c}}$ [see Fig. 5(c)]. Inversely, in-plane resistivity $\rho_{xx}$ presents peaks for fields applied along the hard axis and flat minima for field applied along the diagonal easy axis.

The behavior of the magnetoresistance anisotropy for PCCO is significantly different from YBCO and LSCO. For YBCO, there is a clear sign change when magnetic field is applied parallel and transverse to the applied in-plane current. In LSCO, no sign change, but the presence of four asymmetrical lobes was attributed to twinning. In our case, we never observe a sign change in the MR with angle (it always remains negative) and the oscillations are not sensitive to the direction of the applied current as they remain about the same height for the field parallel [0°] and transverse [90°] to the current, except for the additional features observed at high magnetic fields in $\rho_{xx}$. The magnitude of the in-plane MR oscillations is much smaller for PCCO than for YBCO and LSCO. In both cases however, the oscillations disappear close the non-metal to metal crossover. We suspect that orthorhombic distortions could be a key player in promoting the differences in the magnitude and the anisotropy of these MR oscillations in YBCO, LSCO and PCCO.

MR oscillations of c-axis resistivity in cuprates were first reported in strongly overdoped Tl-based cuprates with $T_c \approx 25K$. In this case, the fourfold oscillations are large (0.33% of the total resistance at 40K and 10T) and correspond to a doping region of the phase diagram where both $\rho_{xx}$ and $\rho_c$ remain metallic-like over the whole temperature range. Because the mean-free path (MFP) is fairly large in this strongly overdoped regime, Dragulescu et al. argued that the oscillations in Tl-based cuprates could be due to angular-dependent magnetoresistance oscillations. In our case, the very small MFP for this doping precludes such interpretation.

Recent experiments indicate that the carriers injected into the cuprates through chemical substitution have a strong tendency to distribute non-uniformly in the copper-oxygen planes, segregating into phase-separated regions, clusters and stripes. Several experimental observations, including the MR oscillations obtained with YBCO and LSCO, fit into this
possible scenario. However, only a recent report by Sun et al. indicates the possible presence of stripes in the electron-doped cuprates \cite{Sun2011}. Assuming their existence in the electron-doped cuprates \cite{Jia2012}, we should expect the c-axis resistivity to decrease whenever stripes in adjacent CuO$_2$ planes are directly on top of each other and aligned in the same direction: in this particular case, electrons can tunnel more easily between planes, thus decreasing resistivity. However, our data show that $\rho_c$ is higher when the field is high (for well aligned domains). We suggest here that a low density of stripes in adjacent planes makes it more difficult to have stripes on top of each other whenever domains are well aligned (high fields), in particular if domains on adjacent planes are not or weakly correlated (as in LSCO \cite{Liu2013}). At low fields, random orientation of stripe domains could lead to a better overlap from weakly correlated adjacent planes, and thus to a better conductivity. For in-plane MR [see Fig. 1(a)], the application of in-plane magnetic field could promote a partial displacement of domain walls (the stripes) \cite{Kamiya2014}, enough to change the resistivity and improve the channeling of electrons along longer conducting ”rivers of charges”. Because the in-plane MR oscillations are so small, the changes in domain wall configurations are probably scarce. Our data would suggest that such modifications of the wall configuration are hard to develop only along the hard axis (the Cu-O bonds), leading to maxima in $\rho_{xx}$.

We should mention that this simple scenario ignores completely the possibility of interaction with the underlying rare earth magnetic order (different in PrCeCuO, NdCeCuO, SmCeCuO) \cite{Yang2015}. Our preliminary data on NdCeCuO and SmCeCuO showed no significant difference for the field and angular dependence of $\rho_{ab}$ and $\rho_c$, thus ruling out a direct implication of rare earth magnetism. Moreover, it remains unclear how an electronic system could present at the same time signatures consistent with 2DWL by disorder and one dimensional features like stripes, unless the spin stripes induce only a partial charge density wave in the CuO$_2$ planes. This aspect will need further exploration, possibly through the doping dependence of the observed oscillations.

In summary, we presented anomalous magnetoresistance oscillations for non-superconducting electron-doped cuprates in their non-metallic regime for magnetic field applied along the copper-oxygen planes. We showed that sharp fourfold oscillations persist over the whole non-metallic regime for both in-plane ($\rho_{xx}$) and out-of-plane ($\rho_c$) resistivities. The field dependence of $\rho_c$ presents irreversibilities which can be explained by the presence of magnetic domains. Their origin is probably related to the presence of stripe domains in
the cuprates.

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