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Transport evidence of a magnetic quantum phase transition in electron-doped high-temperature superconductors

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We present magnetotransport evidence for antiferromagnetism in films of the electron-doped cuprates $Pr_{2-x}Ce_xCuO_4$. Our results show clear signature of static or quasistatic antiferromagnetism up to optimal doping $x=0.15$, with a quantum phase transition close to $x=0.16$, and a coexistence of antiferromagnetism and superconductivity for $0.12 \leqslant x \leqslant 0.15$.

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In strongly correlated electron systems, quantum fluctuations close to a quantum critical point lead to many exotic properties of matter.[1,2] One example is the unconventional superconductivity (SC) and the non-Fermi liquid normal state properties, which appear close to a quantum phase transition (QPT). Such phenomena are found in many heavy Fermion[3,4] and organic[5] superconductors. However, attempts to apply quantum phase transition ideas to describe the properties of the high-T_C cuprate superconductors are controversial. In the hole-doped (p-type) cuprates, whether a superconducting fluctuation scenario[6] or a competing order scenario[7–9] is an appropriate description of the pseudogap phenomena is still highly debated. In the electron-doped (n-type) cuprates, the existence of an antiferromagnetic to paramagnetic QPT is more plausible, but there is significant disagreement over if, and where, it occurs and its role in the physical properties.[10–21]

Several transport studies[10–12] on electron-doped $Pr_{2-x}Ce_xCuO_4$ (PCCO) thin films suggest an antiferromagnetic QPT inside the superconducting dome at $x \approx 0.16$, which is slightly above the optimal doping. Angle resolved photoemission spectroscopy (ARPES) measurements on $Nd_{2-x}Ce_xCuO_4$ (NCCO) (Ref. 18) and optical measurements on NCCO and PCCO (Refs. 19 and 20) revealed a normal-state gap which still exists at the optimal doping $x=0.15$. However, a recent inelastic neutron scattering (INS) measurement on NCCO single crystals suggests that long-range-order antiferromagnetism (LROAF) does not coexist with SC and an antiferromagnetic QPT occurs just before the superconducting dome at $x \approx 0.13$.[21] A recent ARPES work on superconducting $Sm_{2-x}Ce_xCuO_4$ (SCCO) single crystals suggests a short-range-order antiferromagnetism (SROAF) instead at $x=0.14$.[22] In principle, neutron scattering (NS) and μSR could differentiate these different interpretations. But, so far, measurements from different groups are in significant disagreement.[13–16,21,23] The major experimental difficulty is likely caused by a high-temperature oxygen annealing, which is necessary to achieve superconductivity on the n-type cuprates, but also leads to spurious phases[15] or doping inhomogeneity/uncertainty[16] in large crystals. The controversy over the magnetic properties at high dopings, i.e., $x \geqslant 0.13$, leads to question the nature of the QPT proposed by the transport and optical measurements.

In this paper, we present an in-plane angular magnetoresistance (AMR) study of our PCCO thin films. A fourfold oscillation of the AMR, which is caused by the noncollinear antiferromagnetic structure in the n-type cuprate,[24] is used as an indirect method to track the AFM ordering. The onset temperature of the fourfold AMR, T_A, as shown in Fig. 1, coincides with the Néel temperature T_N of nonsuperconducting NCCO single crystals[21,23] at low dopings ($x \leqslant 0.12$) as expected, but deviates from the recent INS measurements[21] of T_N at high dopings ($x \geqslant 0.12$). Interestingly, T_A extrapolates to zero as $x \rightarrow 0.16$, which is consistent with the Hall[10] and the thermopower[12] signature of a quantum phase transition at the same doping. We believe the fourfold AMR is associated with static or quasistatic antiferromagnetism, and therefore a magnetic origin of the quantum phase transition at $x \approx 0.16$ is suggested by our new results. Our phase diagram also suggests a coexistence of static or quasistatic antiferromagnetism and superconductivity in the doping range $0.12 \leqslant x \leqslant 0:15$, which supports that the superconductivity originates from a magnetic mechanism.

Our c-axis oriented PCCO films were prepared by pulsed laser deposition on $SrTiO_3$ or $LaSrGaO_4$ substrates and were

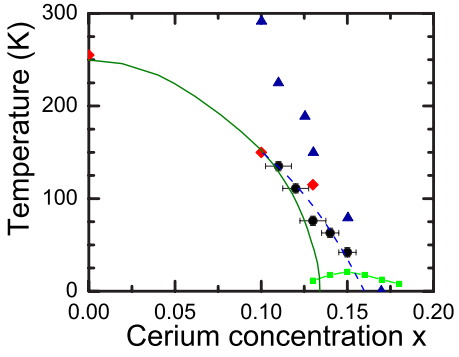

FIG. 1. (Color online) The doping dependence of the onset temperature of static antiferromagnetism, T_A (solid circles), determined from angular magnetoresistance measurements on our $Pr_{2-x}Ce_xCuO_4$ films. The dotted line is a guide to the eye. The Néel temperature T_N of $Nd_{2-x}Ce_xCuO_4$ crystals determined by μSR (Ref. 23) (solid diamonds) and by inelastic neutron scattering (Ref. 21) (solid line), and the normal-state gap onset temperature (solid triangles) determined by the optical measurements (Refs. 19 and 20) are also shown. The solid squares represent the superconducting transition temperature T_C of our films.

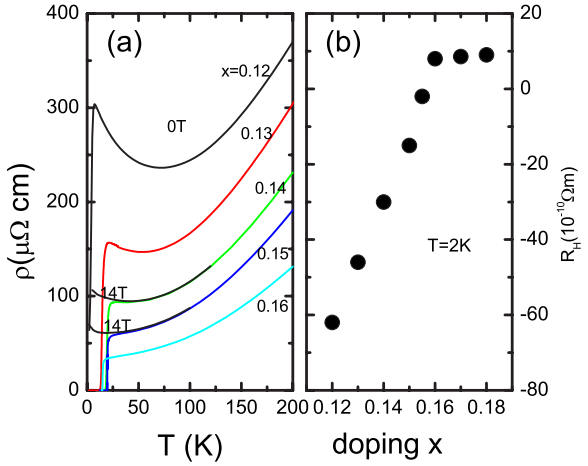

FIG. 2. (Color online) (a) The resistivity of $x=0.12$, 0.13, 0.14, 0.15, and 0.16 films at zero field and at $\mu_0 H=14T\|c$ axis. (b) The Hall coefficient of $0.12\leqslant x\leqslant 0.18$ films ($T=2$ K).

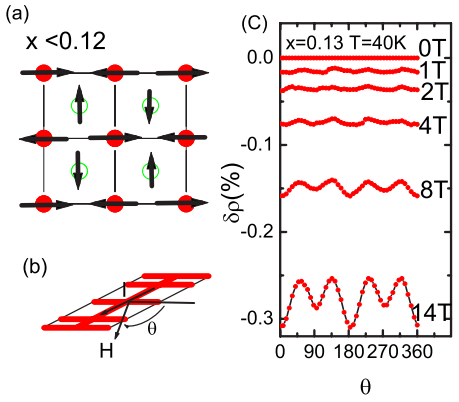

FIG. 3. (Color online) (a) The noncollinear antiferromagnetic structure of the *n*-type cuprates. Solid and hollow circles represent the Cu sites in two adjacent planes, respectively, and arrows indicate the orientation of the magnetic moments (parallel to the lattice *a* axis). (b) The Hall bar pattern of films and the *ab*-plane magnetic field for the angular magnetoresistance (AMR) measurements. θ is defined as the angle between the magnetic field and the Hall bridge (parallel to the lattice *a* axis). (c) The AMR, $\delta\rho(\theta,H)=[\rho(\theta,H)-\rho(\theta,H=0)]/\rho(\theta,H=0)$, for an $x=0.13$ film at different fields at $T=40$ K.

reduced *in situ* under optimized conditions.[25] The superconducting transition temperatures T_C, determined by ac susceptibility, are shown in Fig. 1. All samples are of typical thickness 3000 Å, patterned into a Hall-bar shape, and measured in a quantum design physical property measurement system (PPMS). The resistivity and Hall coefficient at a few dopings are shown in Fig. 2. Two features are clearly seen, both of which have been the subjects of many studies in the literature. First, a low-temperature resistivity upturn occurs in the normal state at low dopings and disappears at $x>0.16$. The antiferromagnetic transition temperature is higher than the upturn temperature at low dopings, but it has been suggested that the upturn is related to the AFM.[11] Second, the low-temperature Hall data show two different doping dependences, which are connected by a kinklike feature at $x=0.16$.[10] The evolution of the Hall data with doping is consistent with a proposed spin-density-wave to paramagnetic QPT at $x\approx 0.16$.[26]

Many underdoped *n*-type cuprates have a noncollinear antiferromagnetic structure[27] below the Néel temperature, as represented in Fig. 3(a). A fourfold AMR of the in-plane transport has been found in highly underdoped, antiferromagnetic $Pr_{1.29}La_{0.7}Ce_{0.01}CuO_4$ crystals.[24] This is caused by a strong spin-orbit coupling and an anisotropic (fourfold) spin-flop field, with the easy-axis along the lattice diagonal direction and the hard-axis along the lattice *a* axis.[24] In this paper, we focus on our in-plane angular magnetoresistance studies on PCCO films, in particular at high dopings. Our films are mounted on a rotator and the sample rotates around the lattice *c* axis with the magnetic field confined in the *ab* plane [see Fig. 3(b)]. The AMR of an $x=0.13$ superconducting film, $\delta\rho(\theta,H)$, is shown at fields up to 14 T at 40 K in Fig. 3(c). With increasing field, a small modulation of the AMR develops. At $\mu_0 H=14$ T, a fourfold oscillation is clearly seen.

A fourfold oscillation is found to exist below a specific temperature in all films with doping up to $x=0.15$. In Figs. 4(a)–4(d), the relative AMR, $\delta\rho'(\theta,14T)$, is plotted to show the temperature dependence of the AMR modulation for several dopings. For each doping, the fourfold oscillation of the AMR emerges below an onset temperature T_A. In Fig. 4(f), the temperature dependence of the magnetoresistance with field along the lattice diagonal direction $\theta=135°$ (easy axis) is plotted. $\delta\rho'(\theta,14T)$ also shows a kinklike behavior at T_A for each doping. The values of T_A for films $0.11\leqslant x\leqslant 0.15$ are plotted in Fig. 1. T_A decreases from 135 K to 65 K as

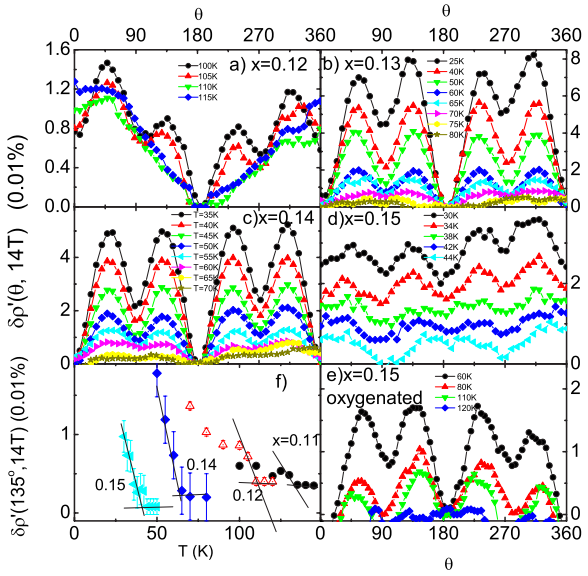

FIG. 4. (Color online) (a)–(e) The temperature dependence of the relative AMR $[\delta\rho'(\theta,14T)=\delta\rho(\theta,14T)-\delta\rho(\theta=180°,14T)]$ for optimally oxygen-reduced $x=0.12$, $x=0.13$, $x=0.14$, and $x=0.15$ PCCO films, and an oxygenated $x=0.15$ PCCO film at 14 T. Plots in (d) are shifted vertically for clarity. (f) The temperature dependence of $\delta\rho'(\theta,14T)$ of several films with the magnetic field along the lattice diagonal direction $\theta=135°$.

doping increases from $x=0.11$ to 0.14. For optimal doping $x=0.15$, a fourfold pattern is also clearly seen at $T=30$ K and not discernible above $T=44$ K. For overdoped $x=0.155$ and $x=0.16$ films (data not shown), the fourfold oscillation is not seen above T_C. At present, we cannot measure AMR in the superconducting samples below T_C since the in-plane H_{C2} is too large. Therefore, if the fourfold oscillation exists, its onset temperature is below T_C.

For comparison, the Néel temperature T_N of nonsuperconducting NCCO crystals from μSR (Ref. 23) and INS (Ref. 21) measurements is also plotted in Fig. 1. We also show the temperature T_P, below which a partial gap in σ_{ab} is seen in the optical measurements.[19,20] The fact that T_P is higher than T_N has been attributed to antiferromagnetic fluctuations.[19,20] For underdoped, nonsuperconducting ($x\leqslant 0.12$) films, T_A is much lower than T_P, but consistent with the value of T_N. In Fig. 4(e), we also show the AMR data for an oxygenated, nonsuperconducting $Pr_{1.85}Ce_{0.15}CuO_4$ film, which has a resistivity similar to as-grown $Pr_{1.85}Ce_{0.15}CuO_4$ crystals. Our low-temperature AMR data is consistent with an earlier AMR study on as-grown $x=0.15$ crystals.[28] Both the amplitude and the onset temperature of the fourfold AMR are much higher than that of the superconducting $x=0.15$ film [Fig. 4(d)]. The fourfold AMR emerges below an onset temperature $T_A \approx 115$ K, again consistent with the Néel temperature determined by elastic neutron scattering of as-grown crystals.[29] *These facts strongly suggest that the fourfold AMR at low dopings is indicative of LROAF, and not antiferromagnetic fluctuations*.

For higher doping $0.13\leqslant x\leqslant 0.15$, no LROAF is found in superconducting NCCO crystals by the INS measurement,[21] which seems to be inconsistent with our finite T_A. We believe our fourfold AMR is not due to the spurious magnetic oxide phase[15] nor doping inhomogeneity induced by oxygen reduction. The large increase under oxygenation of the AMR amplitude and the AMR onset temperature [see Figs. 4(d) and 4(e)], rules out a spurious phase contribution. Compared to bulk NS crystals, our films appear to have a good control of the oxygen as indicated by the sharp superconducting transitions found by the ac susceptibility ($\Delta T_C<1$ K for all films). For our $x\geqslant 0.1$ films, we believe the doping inhomogeneity/uncertainty is below $\Delta x=0.005$ due to our very well controlled growth and oxygen annealing conditions. Interestingly, our T_A extrapolated to zero at $x\approx 0.16$. In comparison with the Hall,[10] thermopower,[12] and ARPES (Ref. 18) measurements, our AMR gives further support for a QPT of magnetic origin at this doping.

In order to understand our data compared with the INS data, we suggest a few possible explanations below.

First, the actual carrier density may be different for the INS single crystals and our AMR thin films, due to different oxygen annealing conditions. Therefore, the fourfold AMR in PCCO could be caused by long-range ordering and the QPT at $x=0.16$ is associated with a LROAF. However, this would indicate a shift of doping by $x\approx 0.03$ between our films and the INS crystals, which seems to be unlikely. The INS suggests no coexistence of LROAF and SC, which seems to be inconsistent with our observation of fourfold AFM up to optimal doping. It is possible that PCCO has a long-range AFM QPT inside the superconducting dome, while this is not the case for NCCO.[30] However, this is unlikely because both systems have a similar AFM transition temperature at low dopings and a Fermi surface topological reconstruction at $x\approx 0.16$.[10,18] We believe that INS and Hall (or thermopower) measurements on the same NCCO single crystal should be able to clarify these two scenarios.

Second, the fourfold AMR method may not be able to distinguish between a long-range-order antiferromagnetism and a *static* short-range-order antiferromagnetism. Considering the disappearance of the LROAF at $x\geqslant 0.13$ as shown by the INS data, our fourfold AMR may suggest a SROAF at higher dopings. Indeed, signatures of SROAF have been shown by INS,[21] although this could arise from oxygen inhomogeneity in the larger crystals used for neutron scattering. Recent ARPES measurements on SCCO single crystals are also suggestive of such a SROAF scenario.[22] Therefore, the disappearance of T_A at $x\approx 0.16$ might suggest that the nature of the QPT at $x\approx 0.16$, as revealed by the Hall, resistivity, and thermopower measurements, is related to a SROAF.

Third, the fourfold AMR could also be caused by a *quasistatic* SROAF, i.e., a fluctuating order, if finite disorder pins the slow fluctuation with a time scale sufficient for transport measurements. This could be the case for the optimal-doped $x=0.15$ films, where a much smaller fourfold AMR is observed [Fig. 4(d)].

Surprisingly, the dramatic change of the Fermi surface at $x\approx 0.16$, as shown by the ARPES (Ref. 18) and the transport[10,12] measurements, suggests that the Fermi surface topological reconstruction is strongly affected by this SROAF rather than the LROAF. For comparison, the Fermi surface evolution is usually associated with a long-range ordering in most magnetic systems.[31] Therefore, our data may suggest a type of QPT with different properties from the one found in the INS experiments.

The short-range-ordering phase seems to be similar to a high-pressure partial-order phase beyond a first-order phase transition in MnSi.[32,33] Unfortunately, the amplitude of the AMR is not simply linear in the amplitude of the order parameter, as indicated by the temperature dependence of the fourfold AMR [see Fig. 4(f)]. Therefore, we are not able to distinguish between a first-order and a second-order phase transition by the AMR measurements. Other scenarios, such as a spin-glass transition[34] or stripes[28] are also possible. Although these phases are supported in some *p*-type cuprates,[9,35] strong evidence for either of these has not been found in the *n*-type cuprates.

Our AMR evidence of static or quasistatic antiferromagnetism with a QPT inside the superconducting dome also provides a framework to understand other experimental results in the *n*-type cuprates. First, our T_A is slightly above the resistivity upturn temperature, which supports the view that the upturn (metal-insulator-like transition) is related to the static AFM,[11] although more studies are required to understand this in detail. Our measurement also suggests a coexistence of SROAF and SC up to optimal doping $x=0.15$. The domelike doping dependence of T_C in the underdoped regime may be naturally explained by a competition between the coexisting SROAF and SC.

In summary, we determined the doping dependence of

static or quasistatic antiferromagnetism in the electron-doped cuprates by an angular magnetoresistance method on $Pr_{2-x}Ce_xCuO_4$ films. Our data give evidence for the existence of intrinsic static antiferromagnetism up to $x=0.15$, which is consistent with the proposed quantum phase transition at $x \approx 0.16$.[10–12] Compared with the inelastic neutron scattering evidence[21] for a long-range-order antiferromagnetic quantum phase transition at $x \approx 0.13$, our angular magnetoresistance measurements suggest a short-range-order antiferromagnetic quantum phase transition at $x \approx 0.16$.

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