Different roles of cerium substitution and oxygen reduction in transport in $Pr_{2-x}Ce_xCuO_4$ thin films

J. Gauthier, S. Gagné, J. Renaud, M.-È. Gosselin, and P. Fournier*
Regroupement québécois sur les matériaux de pointe and Département de Physique, Université de Sherbrooke, Sherbrooke, Québec J1K 2R1, Canada

P. Richard
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02467-3804, USA
(Received 16 November 2006; published 22 January 2007)

Using pulsed-laser ablation with an improved oxygen annealing process and Hall effect measurements, we show that the reduction process needed to induce superconductivity in electron-doped cuprates thin films does not trigger a significant change in carrier concentration (or band filling) contrary to cerium substitution. We show that it has, however, a severe impact on hole-type carrier mobility. This feature is evidenced by focusing on the overdoped regime ($x \geq 0.16$) for which reduction increases the contributions of hole-type quasiparticle excitations to the Hall coefficient without affecting much the contribution from electrons. Since reduction has been also shown recently to provoke a strong suppression of antiferromagnetic order for doping close to optimal, we interpret the strong increase in mobility to result from a decreasing scattering rate related to a decreasing strength of antiferromagnetic correlations. We suggest that delocalization of hole-type carriers with reduction is achieved through the frustration of the antiferromagnetic order of as-grown nonsuperconducting composition by in-plane oxygen vacancies. We propose a comparison of ARPES data for as-grown and reduced $Pr_{2-x}Ce_xCuO_4$ on the overdoped side as a possible experiment to clarify the origin of the hole-type quasiparticles with reduction.

DOI: 10.1103/PhysRevB.75.024424 PACS number(s): 74.62.Dh, 74.25.Fy, 74.25.Jb, 74.78.Bz

One of the persistent mysteries related to the phenomenon of high-temperature superconductivity (HTSC) is the role of the oxygen reduction leading to superconductivity in the electron-doped (*n*-type) cuprates, $RE_{2-x}Ce_xCuO_{4\pm\delta}$ (RE $=$Pr, Nd, Sm, La).[1] The observed changes in oxygen content during this key reduction process, only of the order of $\delta \sim 0.01$ to 0.04 ($\sim 1\%$) for $x=0.15$,[2–4] have a major impact on the electronic properties of the *n*-type cuprates as they drive the materials from a nonsuperconducting high-resistivity material with a low-temperature nonmetallic regime to a low-resistivity bulk superconductor.[5,6] This reduction leads also to a complete suppression of antiferromagnetism for optimal reduction, leaving only short-range order.[7] None of the other high-T_c superconductors possess such a dual doping (oxygen and cerium) dependence of their properties. Considering that some of the most promising theoretical advances on HTSC have been achieved recently with the *n*-type cuprates,[8–11] it becomes important to fully characterize the real relation between doping, carrier concentration, and band filling, a key parameter used in many theories. Moreover, the two degrees of freedom in doping for this family give us a powerful handle on our ability to modify at will their electronic properties and learn more on the possible mechanism leading to superconductivity.

Relying on elastic neutron scattering data on $Nd_{2-x}Ce_xCuO_{4\pm\delta}$ single crystals,[12,13] it is widely believed that reduction results in the removal of interstitial (apical) oxygens which simply adds electrons to the crystal structure and decreases pair-breaking defect scattering enough to favor superconductivity.[14] However, recent reports of infrared crystal-field spectroscopy as a local probe of the oxygen defect structure in this family[15,16] show clear evidence that apical oxygen, when present, *is not removed by reduction* for $x \neq 0$. The authors proposed instead that reduction leads to oxygen vacancies in the CuO_2 planes for $x \geq 0.08$.[15,16] Such a scenario was proposed also by Brinkmann *et al.*[17] to explain the transport properties observed in $Pr_{2-x}Ce_xCuO_{4+\delta}$ single crystals.

To clarify the real effect of oxygen reduction and the possible impact of this newly identified defect structure, we present a systematic study of $Pr_{2-x}Ce_xCuO_{4+\delta}$ (PCCO) epitaxial thin film transport properties as a function of oxygen content for various cerium contents, but *focusing on the overdoped regime*. The overdoped regime is interesting in itself since, within an appropriate window of cerium doping, the Hall coefficient changes sign as a function of temperature,[14,18] consistent with a two-carrier scenario.[17,19–23] By tuning oxygen doping, one might affect unevenly the behavior of these two types of carriers and understand the actual importance of each type for superconductivity. The overdoped regime was used recently by Higgins *et al.*[24] over a limited oxygen content range to study the effect of disorder induced by irradiation.

Studying closely our $x=0.17$ and 0.20 films over an extended oxygen content, we show that they can also be driven from the nonsuperconducting to the superconducting state ($T_c \sim 15$ K) without reaching the maximum T_c of 23 K obtained for optimal carrier doping. This simple observation and the analysis of the temperature dependence of the Hall coefficient as a function of oxygen content *cannot be* explained by simple changes in the carrier concentration and/or the decrease of the scattering rate by impurities. We propose a unifying scenario for the role of reduction taking into account our results together with recent data from neutron

scattering,[7,25] angle-resolved photoemission spectroscopy (ARPES),[26,27] optical spectroscopy,[28] and theoretical models.[8–11] In this scenario, reduction leads to a decreasing quasiparticle scattering rate through the suppression of antiferromagnetic correlations observed as an increase in mobility of hole-type carriers with little changes in carrier concentration.

I. EXPERIMENTAL SETUP AND PROCEDURES

The PCCO thin films used for this study were grown by pulsed-laser ablation deposition (PLD) on $LaAlO_3$ and $SrTiO_3$ as previously described.[29,30] As shown below, we have achieved a great control in oxygen doping required for this study using molecular oxygen (O_2) instead of nitrous oxide (N_2O) (Ref. 29) as the ambient gas during growth and *in situ* postannealing. The pressure of O_2 in the deposition chamber can be stabilized during postannealing to vary the oxygen content of the films: postannealing pressures in the range from $\sim$0.2 mTorr up to 500 Torr, in addition to vacuum annealing ($P<0.1$ mTorr), were used for this study. Since we cannot measure accurately the oxygen content of the films, we index a batch of films grown in identical conditions, but then postannealed *in situ* at *different pressures* with the same temperature and the same duration of annealing. An increasing index from [1] to [14] implies an increasing pressure during postannealing, thus an increasing oxygen content in the films. Additional films grown and annealed under various conditions (e.g., in N_2O) were also prepared for comparison. The AC susceptibility, resistivity, and Hall effect measurements were all done in a Physical Properties Measurement System (PPMS, Quantum Design). X-ray diffraction in the θ–2θ and the ϕ-scan modes on control samples were done in order to ascertain the orientation of the crystallographic axis and to corroborate the value of their c axis with a previous report.[29] All the films were found to be epitaxial and c-axis oriented, in close agreement with Ref. 29.

II. RESULTS AND DISCUSSION

In Figs. 1(a) and 1(b), we present the temperature dependence of the in-plane resistivity for most of the $x=0.17$ samples prepared under O_2. The gradual variation of the resistivity with oxygen doping and the narrow transitions clearly demonstrate our great control of the reduction/oxygenation process using molecular oxygen. One can observe that the nonsuperconducting over-oxygenated samples (like [14]) present resistivity characteristics similar to reduced underdoped $x=0.05$ and 0.10 thin films previously reported[29–31] (see also Fig. 2 below). Removal of oxygen provokes a strong decrease in resistivity and leads ultimately to superconductivity similar to increasing cerium doping for fully reduced films. In Fig. 1(c), we present the transition temperature T_c as a function of the oxygen content index that one can deduce from the resistivity data of Fig. 1(b).[32] In the same figure, we added also the maximum value of T_c $\sim$16 K reached for films grown in N_2O (cross). The maximum $T_c \sim 14$ K for $x=0.17$ grown in O_2 seems to be part of

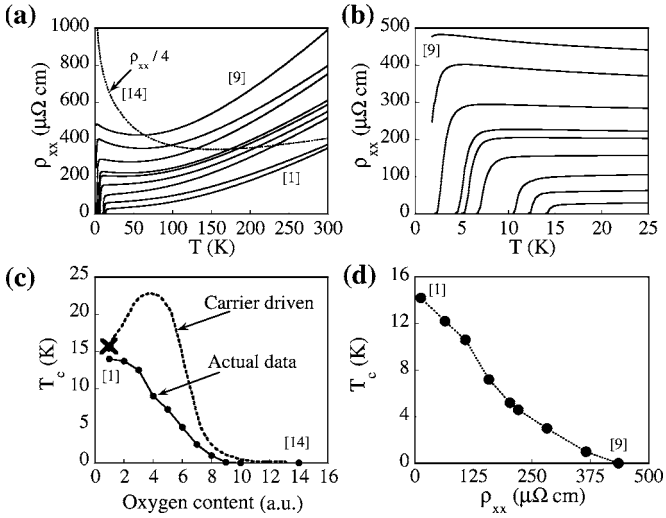

FIG. 1. (a) Resistivity as a function of temperature for $x=0.17$ thin films with various oxygen contents. (b) Low-temperature region of the same data. (c) Critical temperature (T_c) as a function of oxygen content for $x=0.17$ for films grown in oxygen (full circles, solid line is a guide to the eye). Cross: highest T_c under N_2O. Dashed line: schematic of the expected behavior for a carrier driven T_c (see text). (d) T_c as a function of the in-plane resistivity at 30 K.

a plateau, a sign of saturation of the reduction process. This plateau can be confirmed also by changing the other annealing parameters like the temperature, the gas, and its duration. In every case, longer time, higher temperature, and lower oxygen pressure during postannealing always lead ultimately to decomposition instead of an increased T_c for $x=0.17$.

A. Two scenarios

Figure 1(d) presents the direct relation between the resistivity at 30 K and the value of T_c. Two main scenarios have been proposed to explain the changes of resistivity resulting from the reduction process. First, it has been interpreted in recent years as a result of apical oxygen removal. In this scenario, the decreasing content of this *defect* would lead naturally to its decreasing contribution to impurity scattering in the low-temperature resistivity. One would also expect that the removal of apical oxygen should result in carrier doping,[25] leading also to a decrease in resistivity similar to cerium doping. In fact, a combination of both mechanisms could result in a dramatic change as observed experimentally. Moreover, the *apical oxygen scenario* would suggest that reduction leads to a decrease of pair breaking as a mechanism for the onset of superconductivity.[14]

On the other hand, the proposal that oxygen removal in the CuO_2 planes by reduction[15,16] could lead to a similar behavior seems at first glance counterintuitive. Indeed, one expects that oxygen vacancies in the CuO_2 planes should increase dramatically impurity scattering. However, if these defects created in the CuO_2 planes are destroying long-range antiferromagnetism,[7] a sharp decrease in resistivity due to delocalization could overcome the addition of impurity scatterers. Obviously, in this *planar oxygen scenario*, the onset of superconductivity would have nothing to do with a pair-

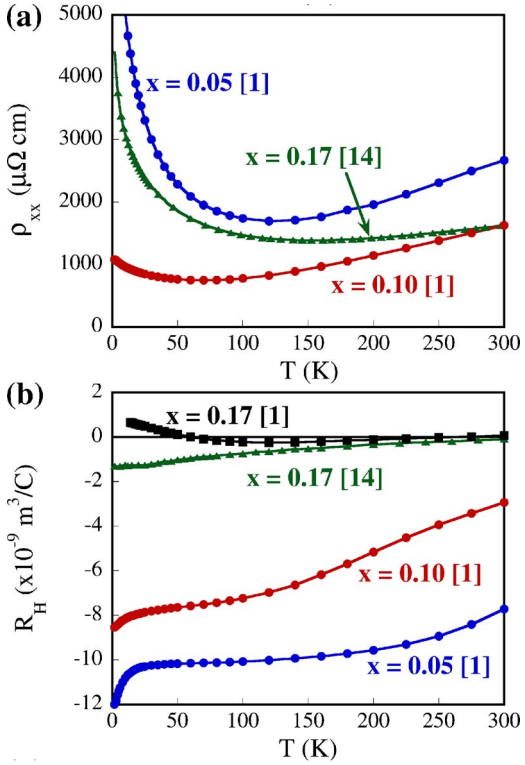

FIG. 2. (Color online) Comparison of (a) resistivity and (b) Hall coefficient temperature dependence for the underdoped reduced $x=0.05$ [1] and 0.10 [1] PCCO thin films, and the reduced $x=0.17$ [1] and overoxygenated $x=0.17$ [14] thin films. The indexes [1] and [14] refer to fully reduced and fully oxygenated films, respectively.

breaking scenario, but rather be consistent with the gradual decrease of the Néel temperature as observed by neutron scattering[25] and the onset of antiferromagnetic fluctuations as a natural possible origin for pairing.[33,34]

B. Hall effect: Little change in carrier concentration

For these two scenarios, the Hall effect is expected to behave quite differently. In the apical oxygen scenario, the decreasing oxygen content should lead to electron doping, similar to increasing the cerium content x,[25] thus to significant changes of the Hall coefficient *at all temperatures* as a function of oxygen content. In the planar oxygen scenario, the two additional electrons resulting from a vacancy would have a strong tendency to localize on the nearby copper sites, giving Cu^{1+} oxidation state. Since these localized electrons could not contribute much to the free carrier concentration in this picture, the Hall coefficient would not depend much on the oxygen content. Thus, a direct examination of the Hall coefficient over an extended range of oxygen content should reveal the actual mechanism leading to the major changes in electronic properties with reduction in the electron-doped cuprates.

Figures 2(a) and 2(b) compare the transport properties of our over-oxygenated PCCO $x=0.17$ [14] and fully reduced $x=0.17$ [1] with the fully reduced $x=0.05$ [1] and 0.10 [1] thin films. Both $x=0.05$ [1] and 0.10 [1] samples should have roughly an amount of oxygen equivalent to the fully superconducting $x=0.17$ [1] since they were reduced in identical conditions. In Fig. 2(a), the resistivity of the $x=0.17$ [14] film is indeed intermediate to the underdoped reduced films. However, the Hall coefficient in Fig. 2(b) is very different. In fact, on first approximation, the magnitude of the Hall coefficient seems to remain directly related to the cerium content x and influenced little by reduction, evidenced by its relatively small variation from $x=0.17$ [1] to [14] with oxygen doping when compared to the large changes provoked by cerium substitution. These data indicate clearly that cerium doping and oxygen reduction do not influence in the same way the physical properties. They suggest instead that while cerium doping controls band filling, reduction affects mainly the mobility of the free carriers.

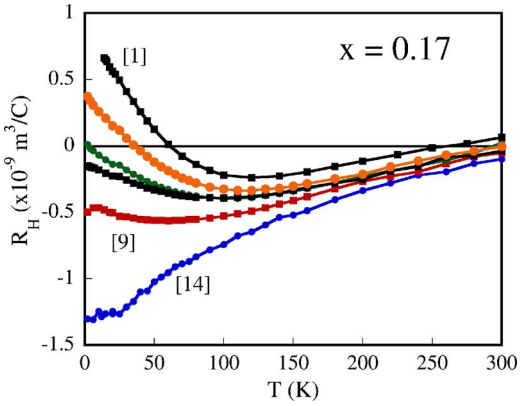

FIG. 3. (Color online) Temperature dependence of the Hall coefficient for various oxygen contents for $x=0.17$. The oxygen content increases from samples [1] to [14].

The role of reduction can be further clarified by measuring the temperature dependence of the Hall coefficient for all of the $x=0.17$ samples shown in Fig. 1. In Fig. 3, a representative sampling of these results is presented. We observe first that the temperature dependence of R_H is strongly affected by the oxygen content *only at low temperatures*, while the progression is relatively modest around room temperature. We should emphasize again that the variations observed at $T=300$ K in Fig. 3 are small when compared to the cerium doping dependence shown in Fig. 2 (see also Ref. 35), clearly underlining the different roles of cerium substitution and oxygen reduction in the doping process. We can control the oxygen content to such a point as to select one leading to a Hall coefficient tending to $R_H=0$ at $T=0$ (as measured in high magnetic field beyond H_{c2}): for $x=0.17$, such a sample has a $T_c \sim 6.5$ K (open squares in Fig. 3).

For $x=0.20$, the variation of the transport properties is very similar, except for a striking difference, as seen in Figs. 4(a) and 4(b). The overoxygenated high-resistivity and nonsuperconducting thin films ($x=0.20$ [14]) present a *fully negative* Hall coefficient for the whole temperature range, while the fully reduced superconducting low-resistivity ones have *only positive* R_H. The magnitude of R_H for the as-grown thin films ($x=0.20$ [14]) shows that the carrier density is *large and negative* demonstrating the dominant role of electron-type carriers. Oxygen reduction provokes a sign change of R_H with hole-type quasiparticles dominating the transport at all temperatures. More importantly, this reduction promotes also superconductivity.

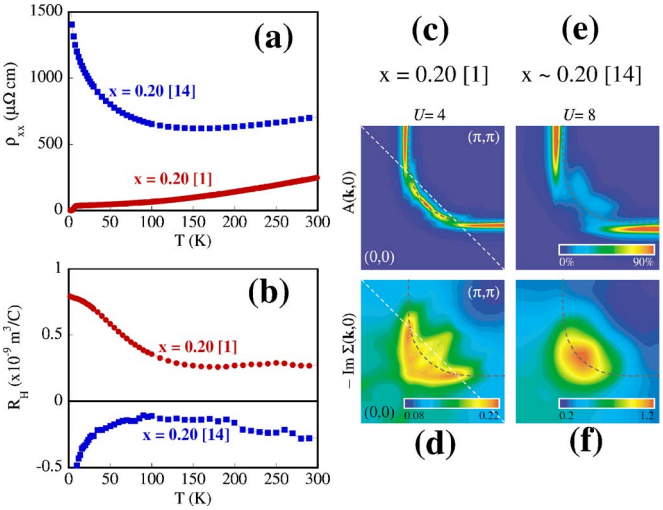

FIG. 4. (Color online) Comparison of (a) resistivity and (b) Hall coefficient temperature dependence for the oxygenated and the reduced PCCO thin films. Full squares and circles are for oxygenated and reduced samples, respectively. Calculated ARPES spectral density $A(\vec{k},\omega=E_F)$ and scattering rate $\Gamma(\vec{k},0)\propto -\mathrm{Im}\,\Sigma(\vec{k},0)$ from Ref. 10 for electron-doped cuprates with band filling of 17% presenting the effect of increasing strength of AF correlations (increasing U). Our reduced samples approach the situation depicted in (c) and (d) while the ARPES response of oxygenated samples should be approaching (e) and (f). See text.

C. Effect on T_c

An important feature of our data is that reduction does not affect the electronic properties in the same manner as the cerium substitution. In Fig. 1(c), a rough schematic of the expected doping dependence of T_c is presented as a dashed line if one assumes that removing oxygen atoms from the structure adds simply (mobile) electrons in the CuO_2 planes and contributes to doping exactly in the same fashion as cerium doping. This scheme assumes that the nonsuperconducting overoxygenated $x=0.17$ PCCO (index = [14]) is very close to the reduced $x=0.05$ PCCO (index = [1]) in terms of electronic properties. From the resistivity data of Fig. 2(a), this assumption looks acceptable. If reduction was only changing the carrier concentration or band filling, we would expect the $x=0.17$ thin films to present a peak in T_c up to $T_{c,\max}\sim 23$ K (the maximum T_c for the PCCO system for $x=0.15$) before going down to 14–15 K as illustrated in Fig. 1(c). Obviously, this is not the case. It is another hint that reduction does not affect the carrier concentration (or band filling) the same way cerium substitution does.

D. Cerium substitution vs oxygen reduction: Two-carrier model

There is a very straightforward observation in our data that deserves more attention. In the case of cerium doping, the changes in R_H and ρ_{xx} are usually large when x is varied as seen in Figs. 2(a) and 2(b).[21,35] However, oxygen reduction has little impact on R_H except at low temperature (see Fig. 3), while it has as large an effect as cerium doping on ρ_{xx}. For instance, the weak oxygen doping dependence of the Hall coefficient at room temperature for the same x value indicates that there is only a small amount of carriers added to the system. However, the corresponding brutal drop of the resistivity at 300 K with decreasing oxygen content indicates that some of the carriers are becoming highly mobile. From this observation, the only possible explanation to such behavior is that reduction affects mainly the mobility of the carriers with little effect on their density, while cerium substitution affects carrier density, thus band filling. Their effects are basically unrelated.

Our Hall effect data indicate that it is possible to tune the sign of the charge carriers using both cerium substitution and oxygen reduction. In some cases, one can generate electron-doped cuprates that present Hall coefficient changing sign as the temperature is lowered. The most obvious and extreme example is the $x=0.17$ [1] sample (Fig. 3) that shows a positive R_H at room temperature, becoming negative at intermediate temperatures, and finally positive at low temperatures. This trend in temperature was underlined in the literature as a good indication that two types of carriers are participating in the transport properties.[17,19–23] In such a case, the Hall coefficient can be expressed in a simplistic Boltzmann transport approach as

$$R_H \approx \frac{pe\mu_p^2 - ne\mu_n^2}{(ne\mu_n + pe\mu_p)^2}, \tag{1}$$

where p and n are, respectively, the hole and electron free carrier density, and μ_p and μ_n are their related mobilities.

One is then tempted to associate the two types of carriers to the different patches (arcs) of the Fermi surface observed by ARPES (Ref. 26) and presented also in Fig. 4(c). At low Ce doping ($x=0.04$ and 0.10 in Ref. 26), corresponding to materials with only negative Hall coefficient [$x=0.05$ [1] and $x=0.10$ [1] in Fig. 2(b)], ARPES observes Fermi energy crossing only close to the $(\pm\pi,0)$ and $(0,\pm\pi)$ saddle points in the Brillouin zone (BZ) [as in Fig. 4(e)]. These states have been associated to the incoherent response observed by Raman scattering.[36] Since the corresponding Fermi arcs evolve monotonically with cerium content, following roughly band filling, we will assume that the character (sign) of these quasiparticles does not change with doping, and that the excitations around the $(\pm\pi,0)$ and $(0,\pm\pi)$ points retain their electron-like character. At larger cerium content ($x\sim 0.15$), ARPES shows new Fermi arcs that are observed close to the $(\pm\pi/2,\pm\pi/2)$ points in the BZ. They are separated from the $(\pm\pi,0)$ and $(0,\pm\pi)$ arcs by the pseudogap (hot spots) at the crossing points with the antiferromagnetic Brillouin zone (AFBZ) and have been associated to the coherent response in Raman scattering.[36] In what follows, we will assume that these portions of the Fermi surface around $(\pm\pi/2,\pm\pi/2)$ give rise to the hole-like quasiparticle excitations, similar to the ones observed in hole-doped cuprates.[37] In essence, these hole-like excitations are responsible for the low-temperature positive upturn in $R_H(T)$ seen in Fig. 3 which, using Eq. (1), implies that the mobility of the hole carriers becomes important at low temperature.

Polarized neutron scattering studies in $Nd_{2-x}Ce_xCuO_4$ (NCCO) show that oxygen reduction results in the decrease

of the Néel temperature (T_N).[7,25] For low Ce doping, reduction has little effect on T_N while it suppresses long-range order at large Ce doping (around $x \sim 0.15$) where superconductivity appears. Mang *et al.* proposed initially that one could use a simple rigid shift of the $T_N(x)$ line to explain their whole dataset in accordance with a simple carrier (electron) doping scenario.[25] Our data, in particular the Hall effect in Figs. 2(b) and 3, discard reduction as a simple carrier doping process and clearly show the unequivalent roles of cerium substitution and oxygen reduction. Together with our transport data, neutron scattering suggests instead a direct relation between the decreasing strength of AF correlations with reduction and the observed increasing mobility of the hole carriers.

From recent numerical simulations on the origin of the normal-state pseudogap using nonperturbative two-particle self-consistent (TPSC) (Ref. 9) and cluster-perturbation theory (CPT) (Ref. 10) approaches, it was concluded that the pseudogap observed by ARPES (Refs. 26 and 27) and optical spectroscopy[28,38] is generated in the presence of short-range correlations with no need for long-range order.[10] As obtained by the CPT approach in Figs. 4(c) to 4(f), the hole-like quasiparticle excitations around $(\pi/2,\pi/2)$ are sensitive to the strength of these AF correlations. As the imaginary part of the self-energy $-\mathrm{Im}\,\Sigma(\vec{k},\omega=0)$ around $(\pi/2,\pi/2)$ grows with stronger correlations [controlled by U in Figs. 4(c) and 4(e)], the $(\pi/2,\pi/2)$ hole arcs are gradually suppressed. In fact, the data in Figs. 4(c) and 4(e) reveal that the changes in scattering rate are large for all $\vec{k}$ on the Fermi surface and not only restricted to the hole pockets (see the different color scales in both figures). This would explain why the mobility of the electrons is also sensitive to the reduction. Our transport data suggest that reduction helps decreasing the scattering rate $[\Gamma(\vec{k},0) \propto -\mathrm{Im}\,\Sigma(\vec{k},0)]$ over the whole Fermi surface, revealing the hole-like $(\pi/2,\pi/2)$ quasiparticle excitations without changing significantly band filling. Such changes in scattering rate for both holes and electron quasiparticle excitations are *perceived* as an increase in their mobility μ_p and μ_n in Eq. (1).

Based on recent infrared studies[15,16] showing that reduction leads to oxygen vacancies in the CuO_2 planes, we suggest that such defects induce a partial frustration of the noncolinear AF order,[39] enough to decrease the strength of AF correlations and driving T_N to zero without changing the density of mobile carriers or band filling. The oxygen vacancies add *localized* electrons (not contributing to free carrier density) on neighboring copper sites first destroying long-range order, then promoting the emergence of hole-like quasiparticles around $(\pi/2,\pi/2)$ as AF correlations are continuously suppressed. In this scenario, the suppression of AF correlations has a major impact on the contribution of both types of carriers to the electronic properties through the decrease of the scattering rate over most of the BZ which is perceived as an enhancement of the free carrier mobility (for both electrons and holes) as seen mainly in the strong variation of the resistivity with oxygen content.

Our data for $x=0.20$ show clearly that reduction is tuning the relative importance of electrons and holes for the determination of the overall physical properties. In Fig. 4(b), R_H changes sign completely with reduction. The samples are nonsuperconducting for high oxygen content with fully negative and quite small $R_H(T)$ (large negative carrier concentration), and high resistivity (low mobility), while it is superconducting for reduced samples with fully positive $R_H(T)$ and low resistivity (high mobility). This is a key observation since it indicates first that it is not sufficient to have a lot of negative carriers to get superconductivity. Actually, it is pointing very strongly toward the *necessity* of having mobile $(\pi/2,\pi/2)$ hole-type carriers in this family, as in the hole-doped cuprates, in order to generate superconductivity.

As a concluding remark, we propose that ARPES on reduced and as-grown (oxygenated) single crystals with doping in the optimal to overdoped range ($x=0.15$ to 0.17) should be investigated thoroughly. It could confirm that reduction suppresses the scattering rate around $(\pi/2,\pi/2)$ leading to a gain in intensity in $A(\vec{k},\omega)$ at $\omega=0$ (at the Fermi energy). Essentially, the $A(\vec{k},\omega)$ ARPES data of the reduced and as-grown samples should look fairly similar qualitatively to the calculations and results shown in Figs. 4(c) and 4(e), respectively.

III. SUMMARY

In summary, we have shown that the role of reduction, unlike cerium doping, in the electron-doped cuprates is *not* to provide additional carriers. We use first the trends of the critical temperature, the resistivity, and the Hall coefficient with oxygen doping to confirm that the maximum T_c that can be reached for a fixed cerium content x is solely determined by x. Using overdoped $Pr_{2-x}Ce_xCuO_4$ thin films with $x=0.17$ and 0.20, we show that reduction increases the mobility of hole-type carriers. Relying on recent numerical calculations, we relate the perceived changes of mobility to a significant change in quasiparticle scattering rate as the antiferromagnetic correlations are tuned by reduction. In particular, hole-like carriers associated with Fermi arcs at $(\pi/2,\pi/2)$ as observed by ARPES are favored when antiferromagnetic correlations are suppressed. We predict that these hole-like quasiparticle arcs seen by ARPES should be strongly suppressed in the as-grown case. We suggest that, contrary to the widely accepted interpretation based on impurity scattering by apical oxygen (defects), oxygen vacancies in the copper-oxygen planes are likely responsible for the decreasing strength of antiferromagnetic correlations and eventually lead to a decrease in scattering rate by antiferromagnetic fluctuations, allowing for the appearance of the hole pockets and ultimately to superconductivity.

ACKNOWLEDGMENTS

We thank R. L. Greene, V. Hankevych, A.-M. Tremblay, D. Sénéchal, G. Riou, M. Poirier, and S. Jandl for several fruitful discussions and S. Pelletier for technical assistance. We acknowledge the support of CIAR, CFI, NSERC (Canada), FQRNT (Québec), and the Foundation of the Université de Sherbrooke.

*Electronic address: Patrick.Fournier@USherbrooke.ca

[1] H. Takagi, S. Uchida, and Y. Tokura, Phys. Rev. Lett. **62**, 1197 (1989).
[2] P. Klamut, J. Alloys Compd. **194**, 15 (1993).
[3] Y. Idemoto and K. Fueki, Jpn. J. Appl. Phys., Suppl. **30**, 2471 (1991).
[4] E. Navarro, D. Jaque, J. Villegas, J. Martyn, A. Serquis, F. Prado, A. Caneiro, and J. Vicent, J. Alloys Compd. **323-324**, 580 (2001).
[5] W. Jiang, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. B **47**, 8151 (1993).
[6] W. Jiang, S. N. Mao, X. X. Xi, X. Jiang, J. L. Peng, T. Venkatesan, C. J. Lobb, and R. L. Greene, Phys. Rev. Lett. **73**, 1291 (1994).
[7] E. M. Motoyama, G. Yu, I. M. Vishik, O. P. Vajk, P. K. Mang, and M. Greven, cond-mat/0609386 (2006).
[8] C. Kusko, R. S. Markiewicz, M. Lindroos, and A. Bansil, Phys. Rev. B **66**, 140513 (2002).
[9] B. Kyung, V. Hankevych, A.-M. Daré, and A. M. S. Tremblay, Phys. Rev. Lett. **93**, 147004 (2004).
[10] D. Sénéchal and A. M. S. Tremblay, Phys. Rev. Lett. **92**, 126401 (2004).
[11] J. Lin and A. J. Millis, Phys. Rev. B **72**, 214506 (2005).
[12] A. J. Schultz, J. D. Jorgensen, J. L. Peng, and R. L. Greene, Phys. Rev. B **53**, 5157 (1996).
[13] P. G. Radaelli, J. D. Jorgensen, A. J. Schultz, J. L. Peng, and R. L. Greene, Phys. Rev. B **49**, 15322 (1994).
[14] X. Q. Xu, S. N. Mao, W. Jiang, J. L. Peng, and R. L. Greene, Phys. Rev. B **53**, 871 (1996).
[15] G. Riou, P. Richard, S. Jandl, M. Poirier, P. Fournier, V. Nekvasil, S. N. Barilo, and L. A. Kurnevich, Phys. Rev. B **69**, 024511 (2004).
[16] P. Richard, G. Riou, I. Hetel, S. Jandl, M. Poirier, and P. Fournier, Phys. Rev. B **70**, 064513 (2004).
[17] M. Brinkmann, H. Bach, and K. Westerholt, Physica C **292**, 104 (1997).
[18] P. Fournier, P. Mohanty, E. Maiser, S. Darzens, T. Venkatesan, C. J. Lobb, G. Czjzek, R. A. Webb, and R. L. Greene, Phys. Rev. Lett. **81**, 4720 (1998).
[19] Z. Z. Wang, T. R. Chien, N. P. Ong, J. M. Tarascon, and E. Wang, Phys. Rev. B **43**, 3020 (1991).
[20] M. A. Crusellas, J. Fontcuberta, S. Pinol, M. Cagigal, and J. L. Vicent, Physica C **210**, 221 (1993).
[21] F. Gollnik and M. Naito, Phys. Rev. B **58**, 11734 (1998).
[22] P. Fournier, X. Jiang, W. Jiang, S. N. Mao, T. Venkatesan, C. J. Lobb, and R. L. Greene, Phys. Rev. B **56**, 14149 (1997).
[23] P. Fournier, E. Maiser, and R. L. Greene, in *The Gap Symmetry and Fluctuations in High-T_c Superconductors*, edited by J. Bok, G. Deutscher, D. Pavuna, and S. Wolf (NATO ASI Series B, 1998), Vol. 371, p. 145.
[24] J. S. Higgins, Y. Dagan, M. C. Barr, B. D. Weaver, and R. L. Greene, Phys. Rev. B **73**, 104510 (2006).
[25] P. K. Mang, O. P. Vajk, A. Arvanitaki, J. W. Lynn, and M. Greven, Phys. Rev. Lett. **93**, 027002 (2004).
[26] N. P. Armitage, F. Ronning, D. H. Lu, C. Kim, A. Damascelli, K. M. Shen, D. L. Feng, H. Eisaki, Z.-X. Shen, P. K. Mang *et al.*, Phys. Rev. Lett. **88**, 257001 (2002).
[27] H. Matsui, K. Terashima, T. Sato, T. Takahashi, S.-C. Wang, H.-B. Yang, H. Ding, T. Uefuji, and K. Yamada, Phys. Rev. Lett. **94**, 047005 (2005).
[28] A. Zimmers, J. M. Tomczak, R. P. S. M. Lobo, N. Bontemps, C. P. Hill, M. C. Barr, Y. Dagan, R. L. Greene, A. J. Millis, and C. C. Homes, Europhys. Lett. **70**, 225 (2005).
[29] E. Maiser, P. Fournier, J. L. Peng, F. M. Araujo-Moreira, T. Venkatesan, R. L. Greene, and G. Czjzek, Physica C **297**, 15 (1998).
[30] J. L. Peng, E. Maiser, T. Venkatesan, R. L. Greene, and G. Czjzek, Phys. Rev. B **55**, R6145 (1997).
[31] P. Fournier, J. Higgins, H. Balci, E. Maiser, C. J. Lobb, and R. L. Greene, Phys. Rev. B **62**, R11993 (2000).
[32] Typically, the maximum of the peak in the imaginary part of AC susceptibility is observed to be close to the temperature of zero resistivity. Thus, our T_c is chosen for $\rho_{xx}=0$.
[33] P. Krotkov and A. V. Chubukov, Phys. Rev. B **74**, 014509 (2006).
[34] B. Kyung, J.-S. Landry, and A.-M. S. Tremblay, Phys. Rev. B **68**, 174502 (2003).
[35] Y. Dagan, M. M. Qazilbash, C. P. Hill, V. N. Kulkarni, and R. L. Greene, Phys. Rev. Lett. **92**, 167001 (2004).
[36] M. M. Qazilbash, A. Koitzsch, B. S. Dennis, A. Gozar, H. Balci, C. A. Kendziora, R. L. Greene, and G. Blumberg, Phys. Rev. B **72**, 214510 (2005).
[37] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. **75**, 473 (2003).
[38] Y. Onose, Y. Taguchi, K. Ishizaka, and Y. Tokura, Phys. Rev. Lett. **87**, 217001 (2001).
[39] I. W. Sumarlin, J. W. Lynn, T. Chattopadhyay, S. N. Barilo, D. I. Zhigunov, and J. L. Peng, Phys. Rev. B **51**, 5824 (1995).