Transport properties of $Nd_{1.85}Ce_{0.15}CuO_{4+\delta}$ crystals before and after reduction

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We report in-plane resistivity and Hall-effect studies on the same $Nd_{1.85}Ce_{0.15}CuO_{4+\delta}$ crystals before and after reduction. A dramatic change in both magnitude and temperature dependence of ρ and R_H has been observed. In particular, the magntiude of R_H at $T \geq 100$ K drops about a factor of 2–3 after the reducing process but its T dependence is not affected, suggesting a large increase of mobile-carrier density. These observations are consistent with the proposal that excess oxygen exists in a prepared sample, which may cause localization of the electrons provided by Ce doping. The anomalous T dependence of R_H in this system is also discussed within several proposed models.

The discovery of the electron-doped high-T_c superconductors $R_{2-x}Ce_xCuO_{4+\delta}$ (R =Nd,Pr,Sm)[1] provides evidence that the CuO_2 planes display a symmetry with respect to electron or hole doping. This has stimulated intensive studies on the electronic transport and spin magnetism in this system.[2,3] One long-standing puzzle of these materials is the critical role played by deoxygenation (reduction) in the occurrence of superconductivity, i.e., a small amount of oxygen (up to ~ 0.05 per unit cell) always has to be removed from as-prepared samples in order to achieve superconductivity with the maximum T_c (about 24 K). It was first thought[1] that as-prepared samples had oxygen stoichiometry O_4 and annealing in a reduced atmosphere resulted in oxygen deficiency ($\delta < 0$) and thus increased the electron carrier density. However, chemical analysis,[4] x-ray-absorption,[5] neutron-scattering,[6] and magnetoresistance[7] studies suggest the existence of *excess* oxygen (i.e., $\delta > 0$) in the as-prepared samples. These interstitial oxygens may cause localization of the electrons provided by the Ce doping so that the superconductivity is prohibited in the as-prepared samples. A recent Hall-effect study[8] on polycrystalline samples also concluded that the reduction process delocalizes the doped charge carriers. Understanding the microscopic effects of the reduction process might help to elucidate the possible mechanism for the superconductivity in the n-type cuprate materials. This is an important problem, since experimental work to data suggests that the n-type cuprates are traditional s-wave BCS superconductors driven by the electron-photon interaction,[9] whereas the p-type cuprates may have a more exotic mechanism for the superconductivity.[10]

In this paper, we present a transport study on the *same* single $Nd_{2-x}Ce_xCuO_{4+\delta}$ crystals before and after annealing in a reduced atmosphere N_2. Our results clearly demonstrate that reduction dramatically changes the in-plane resistivity ρ_{xx} and the Hall coefficient R_H, while the T dependence of R_H at $T \geq 100$ K is essentially independent of the reducing process. In addition, we observe a pronounced peak near $T \approx 75-100$ K in $R_H(T)$ for all reduced superconducting samples. We argue that our observation cannot be accounted for by a small increase of the electron carrier density resulted from a reduction in the oxygen content from 4.0, but it is rather consistent with the proposal of excess oxygen in the as-grown crystals. We will also discuss the strong T dependence of R_H in this n-type system within several proposed models.

The crystals with nominal Ce composition $x = 0.15$ were grown by employing a flux method. Details can be found in a previously published paper.[11] The two plate-shaped thin crystals used for reduction study have dimensions of $1.3 \times 0.8 \times 0.02$ mm^3 and $1.6 \times 0.85 \times 0.019$ mm^3, respectively. Energy dispersive x-ray (EDX) measurements done on crystals prepared in the same way shows that the Ce distribution is uniform. The Ce homogeneity in thin crystals is also revealed by an optical study[12] (combining with EDX) on similar prepared thick crystals, which shows that the Ce distribution is uniform within layers of about $20 \sim 25$ μm. We employed a six-lead configuration for simultaneous measurement of ρ_{xx} and R_H. Good electrical contacts were achieved by soldering thin gold wires onto the crystals with indium-silver alloy. The typical contact resistance is of $2 \sim 3$ Ω. To obtain R_H, we sweep the magnetic field from positive to negative values and extract the slope of the resulting transverse resistance data. The normal-state Hall resistance was linear in field at all temperatures. After the transport measurements were done on the as-grown crystals, they were annealed in flowing nitrogen at about 900 °C for 15 h and then were measured again.

Figure 1 shows the T dependence of the in-plane resistivity ρ_{xx} and Hall coefficient R_H of crystal No. 1 (sample NB) before (i.e., as-grown) and after reduction. The as-grown crystal does not show superconductivity in either resistivity or dc susceptibility measurements. Though ρ_{xx} is metallic like at $T \geq 100$ K, it shows a weak upturn below 75 K with the residual resistivity ratio (RRR), $\rho_{xx}(300\text{ K})/\rho_{xx}(25\text{ K}) \approx 1.5$. We attribute this weak upturn to the localization of conducting electrons in the CuO_2 planes, since we also found a negative magnetoresistance at T below the upturn.[13] The Hall coefficient is negative in the whole temperature range, which is consistent with electron-doping in this system, and its magni-

tude decreases monotonically as T increases. After reduction, the same crystal becomes superconducting at $T_c \approx 24$ K with transition width less than 2 K, as confirmed by dc magnetization. Comparing with the data prior to the reducing process, the resistivity is lower (about 3 times smaller) and the RRR becomes much larger (≈ 6). Even down to T_c, ρ_{xx} is still decreasing with no apparent impurity scattering saturation. The magnitude of R_H is found to be 3 times smaller than that of the as-grown crystal. We notice that a pronounced peak around $T \approx 80$ K emerges in R_H, which is quite different from what is seen in the as-grown sample. Since R_H has the same T dependence for $T \geq 100$ K both before and after the reduction, it is quite reasonable to correlate the change in the magnitude of R_H to the change of mobile carrier concentration. This leads us to conclude that deoxygenation dramatically raises the density of mobile charge carriers and drives the sample more metallic and eventually superconducting. The remarkable change in the T dependence of R_H below 100 K will be discussed later in this paper. In order to see if this change in ρ_{xx} and R_H is typical, we have measured another crystal No. 2 (sample ND) prepared in a similar way. A very similar behavior was observed as shown in Fig. 2. Comparing to sample NB, this crystal has significantly smaller ρ_{xx} and R_H before reduction, possibly due to a slightly different oxygen content or Ce composition. This implies that a reduction study on the *same* crystal is absolutely necessary in order to see intrinsic effects.

One of the major objectives of this work is to understand the role of reduction in the occurrence of superconductivity in the $Nd_{2-x}Ce_xCuO_4$ system (NCCO). It was generally believe that as-prepared samples have oxygen content O_4 and the annealing process in an inert atmosphere removes some oxygen. This treatment would result in an increase of the electron carrier density so that the superconducting state might be favored. However, this too simplified model leaves several major problems unresolved. First, it cannot explain why such a small decrease in oxygen content (~ 0.05 maximum) is so critical for inducing superconductivity. In principle, increasing Ce content should do a similar thing, but this is not found experimentally.[14] Second, this model has great difficulty to explain the existence of long-range antiferromagnetism (AFM) of Cu^{2+} in as-grown samples ($x \sim 0.15$), as observed in neutron scattering,[6] because the added electron from Ce dopant should destroy the AFM if it is shared equally between the four Cu sites in the plaquette. Third, if deoxygenation produces oxygen deficiency (i.e., $\delta < 0$), then the reduced sample should be more disordered compared to the as-grown one. Howev-

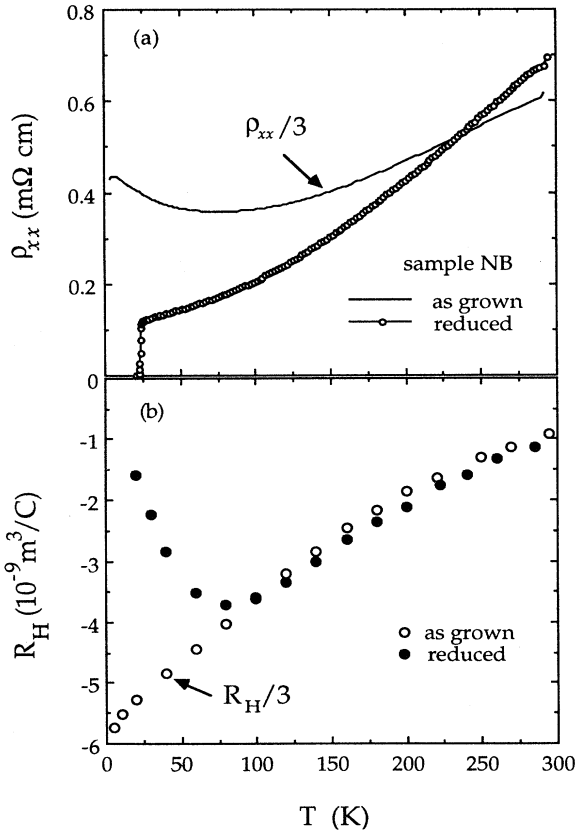

FIG. 1. The T dependence of (a) in-plane resistivity ρ_{xx} and (b) Hall coefficient R_H for crystal No. 1 (sample NB) before (as-grown) and after reduction. The magnitude of ρ_{xx} and R_H for the as-grown crystal is scaled down.

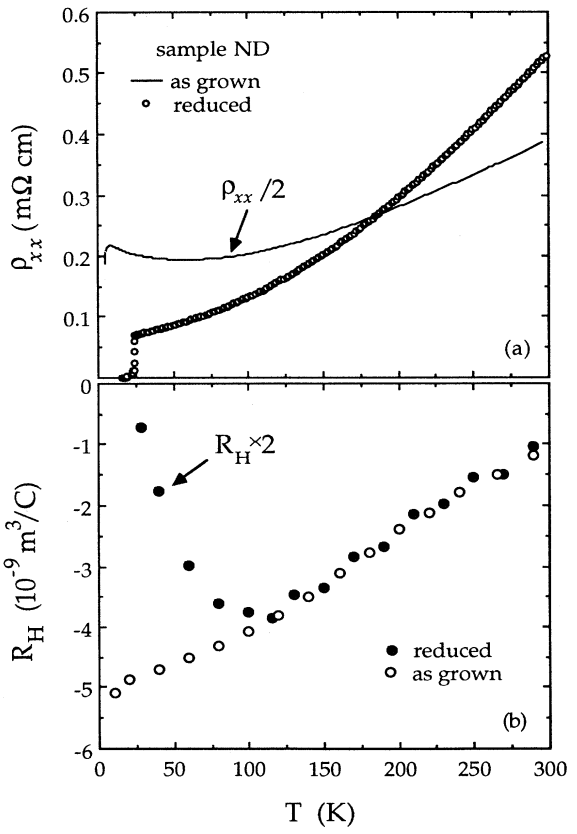

FIG. 2. (a) ρ_{xx} and (b) R_H as the function of temperature for crystal No. 2 (sample ND) before and after reduction.

er, our results show that the opposite case is true: It is the as-grown samples that show localization behavior at low T. Finally, this model cannot explain the drastic change (a factor of 3) in the magnitude of R_H that we observed (see below). Clearly, a more novel mechanism seems to be required.

An early x-ray-absorption study[5] on ceramic samples indicated that the reduction process strongly enhanced the electron density at the Cu sites. This was attributed to the removal of a small amount of defect oxygen atoms at apical sites in the as-grown sample. Matsuda *et al.*[6] also proposed that there is excess oxygen in as-grown $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($\delta>0$) crystals based on their neutron-scattering data. These excess oxygens tend to occupy apical oxygen sites and induce a local disordered potential around them. Since the local square symmetry of Cu ions about the Ce dopant is broken, the electrons provided by Ce doping would tend to be predominantly localized at Cu sites and thus Cu^{2+} antiferromagnetism can still survive at relatively high temperature, consistent with a simple site-dilution model.[6] The deoxygenation process removes these interstitial oxygens and restores the symmetry about the Cu plaquette. In such a situation, electrons will not be localized and thus the Cu^{2+} magnetic correlations will diminish and superconductivity becomes possible. Our transport results are basically consistent with this picture. Specifically, our Hall-effect data on the same crystals before and after reduction gives a strong support to this scenario. In a simplified quasiparticle picture, $R_H \sim 1/ne$, n is the density of charge carriers, and Ce doping will add $2x$ electrons to the unit cell in $Nd_{2-x}Ce_xCuO_{4+\delta}$. In the excess oxygen model ($\delta>0$), the number of carriers per unit cell is $2(x-2\delta)$ (if each interstitial oxygen atom tends to localize two electrons) before reduction and $2x$ after reduction. This gives the ratio of $n_{\text{after}}/n_{\text{before}}=x/(x-2\delta)$. For $x=0.15$ and $\delta=0.05$, this ratio is just 3, consistent with Fig. 1. On the other hand, if the as-grown crystal has oxygen O_4, this ratio will be $(x+2\delta)/x$, which is about 1.7 for the same values of x and δ. We note that this ratio is sample dependent due to the variations of x and δ. Furthermore, since R_H may vary faster than $1/x$ in the superconducting concentration range, caution should be taken when using the relation $R_H \sim 1/ne$.

Further evidence for the existence of excess oxygen, which may localize the conducting electrons at T below ~ 70 K in as-grown samples, comes from our magnetoresistance measurements. In the as-grown crystals, a weak upturn in ρ_{xx} is always seen at low T and the magnetoresistance (MR) in the upturn range shows a negative sign when H is parallel to the c axis (Fig. 3), consistent with a localization behavior. In the superconducting samples, however, this upturn disappears completely and positive MR were observed in the normal state. Uji and Aoki[7] recently carried out a careful MR study in NCCO crystals. They also found that MR is negative and anisotropic in the as-grown crystals at low T, which strongly suggests localization of conducting electrons by a disordered potential possibly created by excess oxygen.

The large increase of the electron carrier density after deoxygenation could also possibly be explained in terms

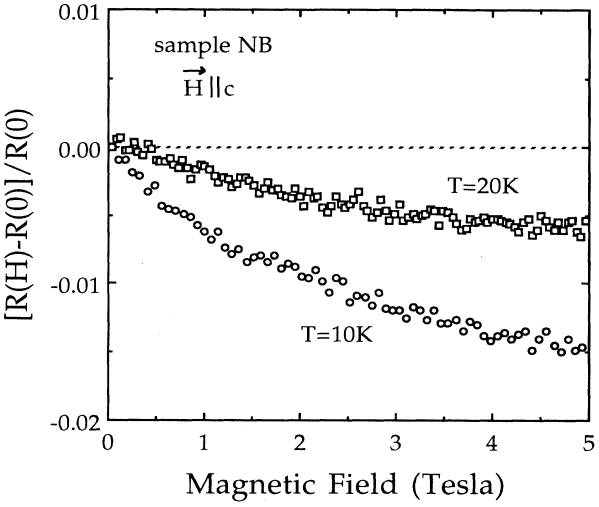

FIG. 3. Magnetoresistance of sample NB (before reduction) at $T=10$ and 20 K.

of the electron neutrality condition for the system. In this approach, reduced samples could have more electrons than as-grown samples based on the requirement of charge compensation due to the removal of the interstitial oxygens. However, there are some difficulties with this electron filling picture. First, the resistivity of the as-grown $x\sim 0.15$ NCCO crystals is much smaller than that of the $x\sim 0.11$ NCCO crystal,[18] but this picture indicates that the former should have a resistivity value similar to that of a $x\sim 0.05$ NCCO crystal for $\delta\sim 0.05$; second, this model cannot explain why only deoxygenation and not increased Ce concentration produces the superconductivity. Therefore, we think the delocalization mechanism may be more consistent with all of the experimental data.

The strong T dependence of Hall coefficient R_H has been observed in most high-T_c cuprates.[15] In $YBa_2Cu_3O_7$ (YBCO) and related systems, the $1/T$ dependence in the normal state was widely observed and has been studied intensively.[16] However, the T dependent R_H of the $Nd_{2-x}Ce_xCuO_4$ system has been investigated far less. There even exists a controversy over the sign of R_H (which is crucial in determining the nature of charge carriers) in this system. Early measurements on ceramics and crystals[1,17,18] revealed electronlike carriers, while one Hall study[19] on crystals showed positive R_H (at least when $T<80$ K). In order to get deeper insight into this discrepancy, we have measured more than ten superconducting crystals (all have thickness within ~ 25 μm in order to have uniform Ce distribution as we reasoned above) with nominal Ce composition $x=0.15$. We should mention that our criterion for a high-quality crystal is based on a sharp transition with a high T_c in both dc susceptibility and resistivity, and a large RRR with no upturn shown in ρ_{xx} at low T. In all high-quality crystals, we found that R_H is consistently negative above T_c with a peak around 75–100 K as shown in Fig. 4. We also observed in one crystal that R_H changes sign from

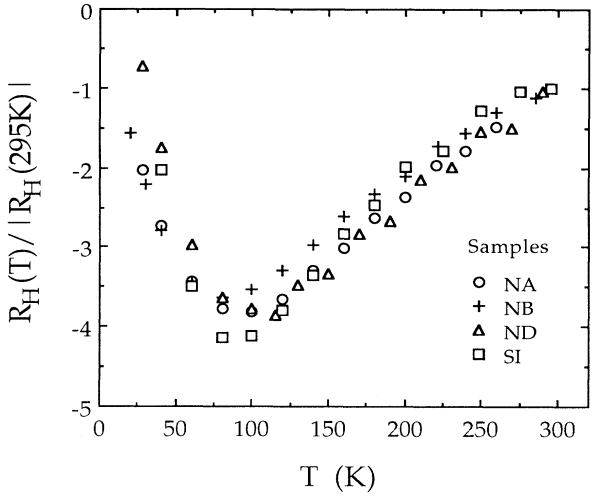

FIG. 4. Normalized Hall coefficient $R_H(T)/|R_H(295\ \mathrm{K})|$ vs temperature for four superconducting NCCO crystals ($x\sim0.15$).

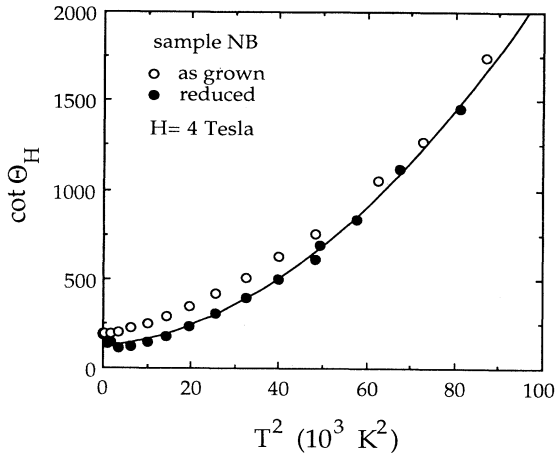

FIG. 5. The cotangent Hall angle ($=\sigma/\sigma_H$) vs T^2 for the same crystal as shown in Fig. 1. The solid line is the best fit to $a+bT^2+CT^4$.

negative to positive at about $T\sim60$ K. We attribute this sign change in R_H to the slightly higher Ce content in this crystal, since R_H develops gradually from negative to positive as x increases.[18,20] At optimum doping ($x\sim0.15$), however, R_H should be negative in the normal state consistent with electron doping.

The most striking feature of $R_H(T)$ in the superconducting NCCO ($x\sim0.15$) crystals is the peak around 75–100 K as shown in Fig. 4. This peak is observed, not only in all of our high-quality crystals, but also in thin films and crystals by other groups.[20,21] Recently, the same feature of R_H has been seen in our NCCO films ($T_c\approx21$ K) as well.[22] Since R_H is positive in the overdoped range ($x\sim0.2$),[18] one might argue that a nonuniform distribution of Ce could cause such a peak. However, we think this is highly unlikely because R_H in the same crystal before annealing shows no anomaly. Also we notice the strong similarity in the T dependence of R_H in NCCO and the $Nd_2CuO_{4-x}F_x$ system.[23] In the latter system, there is presumably no Ce segregation problem. One can also rule out the possibility of oxygen inhomogeneity as the cause for the peak because it is believed that thin films have uniform oxygen distribution. Therefore, we tend to believe this strong T dependence of R_H is intrinsic to the NCCO system for $x\approx0.15$.

Anderson[24] recently proposed a two-dimensional Luttinger-liquid model to explain the T dependence of in-plane ρ and R_H in YBCO by distinguishing transport relaxation rates associated with spinons displaced normal (τ_{tr}^{-1}) and parallel (τ_{cyc}^{-1}) to the Fermi surface. In a simplified quasiparticle picture, the cotangent Hall angle is given by $\cot\Theta_H=\sigma/\sigma_H\sim1/(\omega\tau_{cyc})$, where σ and σ_H are the longitudinal and Hall conductivity, respectively. In YBCO, Pr-YBCO, and $La_{2-x}Sr_xCuO_4$, $\cot\Theta_H\sim AT^2+B$, which was interpreted as the T dependence of the intrinsic scattering rate of spinons according to Anderson's model.[25–27] Similar T dependence of $\cot\Theta_H$ was observed in $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2:2:0:1) and $TlSr_2CaCu_2O_{7-\delta}$ (Tl-1:2:1:2) as well.[28] The quantity $\cot\Theta_H$ should reflect the T dependent $1/\tau_{cyc}$ for a single-band system. In Fig. 5 we plot $\cot\Theta_H(\sim1/\tau_{cyc})$ vs T^2 for sample NB, which clearly shows that $1/\tau_{cyc}$ rises much faster than T^2. Similar behaviors were observed in other crystals as well. This anomalous T dependence does not depend on whether the sample is superconducting or not, so the spinon-spinon scattering idea does not seem to work in the NCCO system. On the other hand, it is expected that $\tau_{tr}\sim\tau_{cyc}$ in a conventional metal. If NCCO behaves like a conventional metal with a single conduction band, $1/\tau_{cyc}$ should follow a nearly T^2 dependence as has been observed in the ρ vs T profile.[29] This is also in disagreement with Fig. 5. A more sophisticated model seems to be required.

It is interesting to compare the present transport data of NCCO with that of the oxygen-doped Tl 2:2:0:1 and Tl 1:2:1:2 systems. In the latter materials, increasing the oxygen content reduces T_c and continuously changes the T dependence of the in-plane ρ from linear to a T^2 behavior, while the T^2 dependence of the Hall mobility μ_H^{-1} ($\sim\cot\Theta_H$) persists. It was thus suggested that the carrier density varies with temperature and a single scattering mechanism ($\sim T^2$) governs the transport properties.[28] This scenario apparently does not apply to the NCCO system, even though NCCO shares some common features in the ρ vs T profile with the oxygen-doped Tl 2:2:0:1 crystals of similar T_c (e.g., sample B in Ref. 28).

A multiband model was also proposed to explain the transport properties of NCCO.[8,19] In a general two-band picture, we have

$$\sigma=\sigma_1+\sigma_2\ , \tag{1}$$

$$R_H=(\sigma_{H1}+\sigma_{H2})/B(\sigma_1+\sigma_2)^2\ , \tag{2}$$

$$\cot\Theta_H=(\sigma_1+\sigma_2)/(\sigma_{H1}+\sigma_{H2})\ , \tag{3}$$

where σ_i and σ_{Hi} represent, respectively, the diagonal and Hall conductivity elements of band i and Θ_H is the Hall angle. The appearance of the peak in $R_H(T)$ could

be naturally understood if the electronic structure of NCCO consists of an electronlike band (band 1, say) and a holelike band. In this case, $\sigma_{H1}<0$ and it should dominate in R_H in order to have the negative sign for R_H in the normal state. The two-band model also seems to be able to explain the discrepancy over the sign of R_H and the Seebeck coefficient in NCCO.[30] However, considering $\sigma\sim\tau_{tr}$, $\sigma_H\sim\tau_{tr}\tau_{cyc}$, and $\rho=1/(\sigma_1+\sigma_2)\sim T^2$, this model does not provide an interpretation for the strong T dependence of $\cot\Theta_H$ as shown in Fig. 5, unless an anomalous scattering process ($\sim T^4$) contributes to $1/\tau_{cyc}$, but not to $1/\tau_{tr}$. This is unlikely for the hole band, since $1/\tau_{cyc}\sim T^2$ has been observed in the p-type YBCO and $La_{2-x}Sr_xCuO_4$ systems.[25–27] It is also highly suspicious that the electron band should possess such a very unusual property. Further work is needed to clarify the origin of this anomalous T dependence of $\cot\Theta_H$ in the NCCO system.

In summary, we have studied the in-plane resistivity and Hall effect in the *same* $Nd_{2-x}Ce_xCuO_4$ crystals ($x\sim0.15$) before and after deoxygenation. We found that the reduction process dramatically changes both the magnitude and T dependence of the resistivity and the Hall coefficient. In particular, a pronounced peak in $R_H(T)$ was observed in all reduced superconducting crystals, which may be closely related to the appearance of superconductivity. The T dependence of R_H at $T\geq100$ K is found to be independent of reduction but its magnitude changes by a factor of $2\sim3$, suggesting a remarkable change in the density of mobile-charge carriers by removing a small amount of oxygen. Our results are consistent with the proposal that the existence of excess oxygen in as-prepared samples causes the localization of the electrons provided by Ce doping. The principal role of deoxygenation then is to remove these excess oxygens and the associated disordered potential so that superconductivity becomes possible. We also found that the cotangent Hall angle in the $Nd_{1.85}Ce_{0.15}CuO_{4+\delta}$ varies much faster than T^2, which cannot be understood within several proposed models for the electronic transport in high-T_c cuprates.

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