Thermomagnetic transport properties of $Nd_{1.85}Ce_{0.15}CuO_{4+\delta}$ films: Evidence for two types of charge carriers

P. Fournier, X. Jiang,* W. Jiang,† S. N. Mao,‡ T. Venkatesan, C. J. Lobb, and R. L. Greene
Center for Superconductivity Research, Department of Physics, University of Maryland at College Park, College Park, Maryland 20742
(Received 9 July 1997)

We report measurements of the thermomagnetic properties of $Nd_{1.85}Ce_{0.15}CuO_{4\pm\delta}$ thin films as a function of oxygen content. The variation of the Nernst coefficient with carrier doping—and its large magnitude—cannot be explained by a conventional single-carrier model. These measurements along with the variation of the Hall coefficient and the thermoelectric power with oxygen doping suggest that a two-carrier model might be suitable to describe the transport properties of these *electron-doped* cuprate superconductors. The ratio of the electrical and thermal Hall mobilities is presented as further evidence for the applicability of a two-carrier model. We finally show the correlation between the doping dependence of the Nernst coefficient and the magnetoresistance. [S0163-1829(97)08645-1]

I. INTRODUCTION

Among the now large family of high-temperature superconducting cuprates (HTSC's), $R_{2-x}Ce_xCuO_{4\pm\delta}$ (R=Nd, Pr,Sm) is one of the rare materials showing a clear n-type Hall coefficient through most of the doping range.[1] This family of materials presents a very distinct set of properties with respect to the other cuprates.[2] The doping interval where a superconducting transition can be observed is smaller by approximately a factor of 3 if a one to one correspondence between the Ce and carrier concentrations is assumed. The actual dependence of the superconducting transition temperature (T_c) as a function of the Ce content looks like a sharp peak, reaching a maximum of approximately 22–24 K at the optimal doping of $x\approx 0.15$ in thin films,[3,4] rather than the typical paraboliclike dependence found in the hole-doped cuprates.[5] The in-plane resistivity (ρ_{ab}) of the superconducting *hole*-doped cuprates has a linear temperature dependence for the underdoped and optimally doped materials,[6] while a nearly T^2 dependence of ρ_{ab} is found for $x\geqslant 0.13$ in $Nd_{2-x}Ce_xCuO_{4\pm\delta}$ (NCCO),[1,7] i.e., for the whole doping range corresponding to the metalliclike materials. The thermal conductivity below T_c of optimally doped NCCO ($x=0.15$) (Ref. 8) does not show the very distinct maximum at approximately 0.5 T_c as observed in $YBa_2Cu_3O_7$ (YBCO) (Ref. 9) and $Bi_2Sr_2CaCu_2O_8$ (BSCCO).[10] In the superconducting state, the temperature dependence of the penetration depth from microwave measurements on the optimally doped NCCO ($x=0.15$) indicates that it is fully gapped[11] (thus, more conventional) and unlikely to present any nodes in the superconducting wave function (or large in-plane anisotropy) as is observed in hole-doped YBCO and BSCCO. Understanding the nature of the differences between NCCO and the other cuprates thus represents a challenge, and might be crucial in clarifying the mechanism of superconductivity in the HTSC's.

It was shown previously that the magnitude and the sign of the normal-state Hall coefficient, R_H, as well as the T_c of NCCO can be varied by changing the oxygen content.[12,13] When comparing the temperature dependence of the Hall coefficients of oxygen-rich, nonsuperconducting $x=0.15$ crystals, and argon-annealed, superconducting ones, the appearance of superconductivity could be correlated with a clear additional *positive* low-temperature component to R_H, pointing out the possibility of holelike charge carriers in superconducting NCCO.[12,14] This is illustrated in Fig. 1. Measurements of the magnetoresistance at 60 K (far enough above T_c to avoid fluctuation effects) show that it becomes positive only in the oxygen doping range where the material is superconducting.[12] Some contradictory reports on the thermoelectric power (TEP) of the optimal NCCO show positive and negative[15] coefficient for all temperatures. In this paper we try to clarify the possible origin of this disagreement.

To probe the apparent contributions of two types of carriers in NCCO, we focus on the nature of the transport properties of thin films with $x=0.15$ as a function of oxygen content. We systematically measure their normal-state Nernst and Hall coefficients, together with the thermoelectric power and the resistivity. We show that the Nernst effect has an unexpectedly large magnitude. Using the ratio of the Nernst coefficient to the thermoelectric power, defined here as the *Nernst* mobility, and comparing it to the Hall mobility, we find that the Nernst mobility is always at least one order of magnitude larger than the Hall mobility. We interpret this

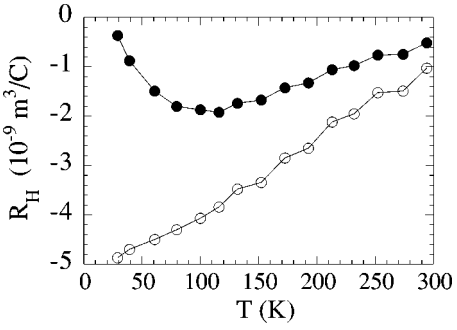

FIG. 1. Hall coefficient as a function of temperature for an as-grown, nonsuperconducting thin film (open circles) and a reduced, superconducting one (solid circles). Solid lines are guides to the eye.

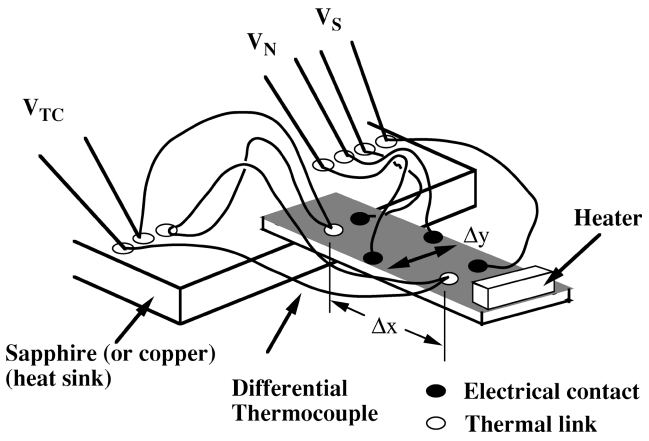

FIG. 2. Schematic of the experimental setup used for the measurement of the thermomagnetic properties.

large inequality as a further indication of two-carrier transport in NCCO.

II. EXPERIMENTAL SETUP AND PROCEDURES

In order to study the pure *ab*-plane components of the thermomagnetic properties, we use thin films, as they are likely to be free of the inhomogeneity problems encountered with polycrystalline samples[16] and single crystals.[17] In Ref. 16, even annealing in equilibrium conditions did not allow the authors to get NCCO polycrystals ($x=0.15$) with a *clear positive slope* ($d\rho/dT>0$) for the resistivity, indicating the difficulty of getting a homogeneous composition throughout polycrystals (including the grain boundaries). Even though single crystals seem to be of reasonable quality as judged by the sharpness of their superconducting transition from in-plane resistivity, dc and ac susceptibility ($\Delta T_c<1$ K), we find that the *c*-axis resistivity of crystals thicker than 20 μm often indicates a wider T_c distribution suggesting Ce (Ref. 18) or oxygen inhomogeneity along the *c* axis.

Our *c*-axis epitaxial thin films for $x=0.15$ are grown by pulsed-laser deposition and are post-annealed following the procedures described elsewhere.[12,19] To summarize our work, the results from four 2000 Å thin films with different T_c's are presented: RED, reduced ($T_c=14.3$ K), OPT, optimal ($T_c=22.5$ K) , OX, oxygenated ($T_c=12$ K), and SOX, superoxygenated ($T_c=0$ K). The films are characterized by high-resolution transmission electron microscopy, wave-dispersive x-ray spectroscopy, and x-ray diffraction showing them to be predominantly *c*-axis oriented, with a small percentage ($<2\%$) of misoriented (110) crystallites.[20]

Details on the thermoelectric power (TEP) and the Nernst effect measurements can be found elsewhere.[21] As illustrated in Fig. 2, a thin film is epoxied onto a copper mount (heat sink) on one side while a small thin-film heater is attached to the other side. A typical longitudinal steady temperature gradient ($\nabla_x T)\leqslant 0.2$ K/mm is applied to the sample. The TEP (longitudinal) and the Nernst (transverse) voltages, respectively, $V_x=E_x(\Delta x)$ and $V_y=E_y(\Delta y)$, are measured with precalibrated gold wires attached onto the film. The thermoelectric power coefficient is then given by $S=E_x/\nabla_x T$, while the Nernst coefficient is $Q=E_y/(B\nabla_x T)$. The thin-film geometry eliminates the contribution arising from the Righi-Leduc effect while measuring the Nernst effect, as the corresponding field-induced transverse temperature gradient is expected to be shorted by the substrate.

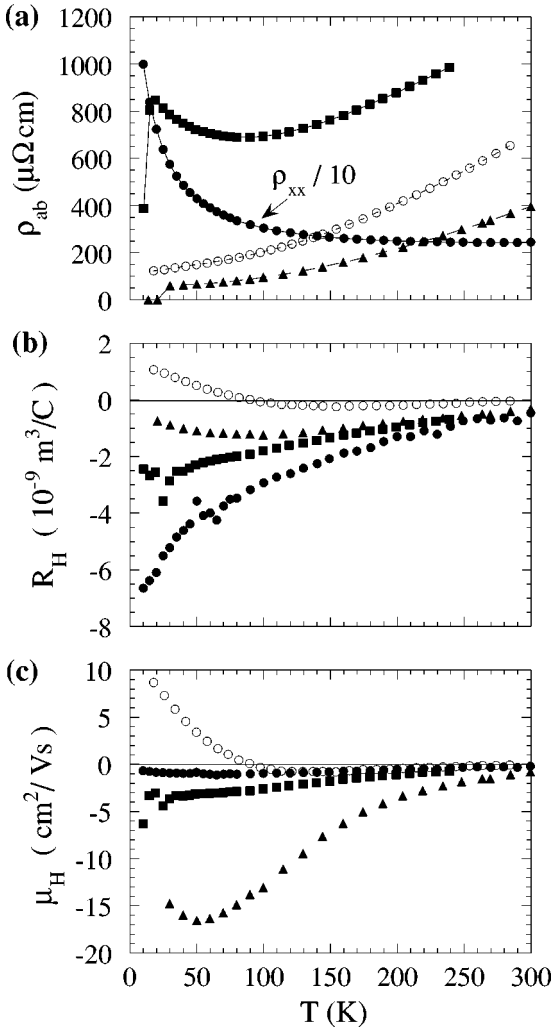

FIG. 3. (a) Resistivity, (b) Hall coefficient and (c) Hall mobility as a function of temperature for all the samples. SOX, solid circles; OX, solid squares; OPT, solid triangles; RED, open circles. Solid lines are guides to the eye.

III. EXPERIMENTAL RESULTS

A. Resistivity and Hall effect

In Fig. 3, we present the in-plane resistivity (ρ_{ab}), the Hall coefficient (R_H), and the Hall mobility ($\mu_H\equiv R_H/\rho_{ab}$) as a function of temperature for all the films. Except for SOX, the in-plane resistivity in Fig. 3(a) is metallicklike (close to quadratic) with a clear positive curvature for most of the temperature range: this is also the case for OX above the upturn temperature. These results are in close agreement with extensive studies of the electrical transport as a function of oxygen content.[12,22]

In Fig. 3(b), we show the temperature dependence of the Hall coefficient for all the thin films. First, we want to emphasize that the magnitude and sign of the Hall coefficient are smoothly varying with oxygen content. The Hall effect is negative at high temperatures for all the samples, increasing slowly in magnitude as the temperature decreases. This is

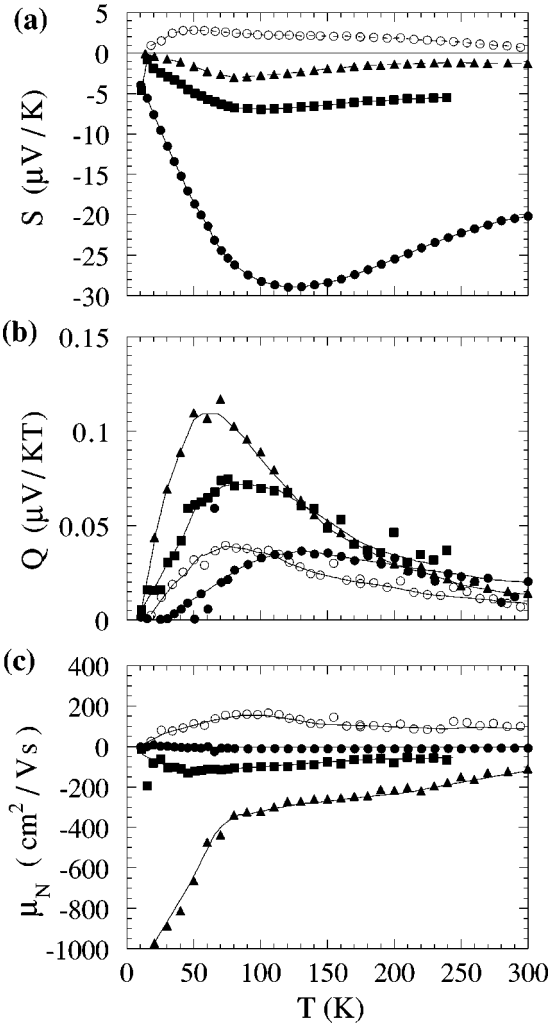

FIG. 4. (a) Thermoelectric power, (b) Nernst coefficient, and (c) Nernst mobility as a function of temperature for all the samples. SOX, solid circles; OX, solid squares; OPT, solid triangles; RED, open circles. Solid lines are guides to the eye.

followed at low temperatures by an additional *positive* contribution starting at around 100 K for the optimally doped films, in agreement with our previous results described in Ref. 12.

In Fig. 3(c), the Hall mobility reveals a continuous variation from a small negative value (SOX) up to a very large negative one (OPT), and finally through an abrupt change of sign to a positive value upon further reduction (RED), resulting from the increasing positive component with *electron* doping. A more thorough analysis of this positive component as a function of doping is likely to shed some light on its origin, but this is beyond the scope of this paper.

B. Thermoelectric power and Nernst effect

Figure 4 shows the thermoelectric power, the Nernst coefficient, and the Nernst mobility $[\mu_N \equiv Q(T)/S(T)]$ for the same films. We first observe that the measured properties appear to vary smoothly with oxygen content. An interesting feature of the TEP in these materials is the fact that electron doping (decreasing oxygen content) leads to a sign change from negative to positive. This is also correlated to the progression of the Hall coefficient with electron doping. In every film, the TEP reaches a maximum absolute value at a concentration-dependent temperature ($T_{\max}$): $T_{\max}$ increases as the oxygen content is increased.

Our results help to explain some of the prior controversy surrounding the sign of the thermoelectric power in these materials.[15] From Fig. 4(a), it is clear that NCCO with $x=0.15$ can potentially show both negative and positive TEP, depending on the oxygen content. However, we do not observe the usual sign change at intermediate temperatures reported for crystals and polycrystals.[15] It is likely that an inhomogenous oxygen content from grain to grain in a polycrystal, as observed recently by Singh *et al.*[23] using high-resolution SEM, could lead to a large variation in the measured TEP above T_c. Even though the main volume of the sample can be off-stoichiometric, an optimum (maximum) T_c could still be observed due to some percolative paths from the properly reduced parts of the sample. Thus, we suggest that inhomogeneities, in particular of the oxygen content, are likely to be responsible for the wide range of TEP observed for polycrystalline NCCO with $x=0.15$.

In Fig. 4(b), we show the temperature dependence of the Nernst coefficient for our samples. For all oxygen contents, the Nernst coefficient remains positive for the whole temperature range. At high temperatures, it seems to follow a T^{-n} relation with $n>1$. It then reaches a maximum at intermediate temperatures before gradually decreasing at low temperatures. Interestingly, the largest Nernst effect is obtained in the optimally doped thin films, in coincidence with the maximum in the Hall mobility.

In Fig. 4(c), we present the Nernst mobility $[\mu_N = Q(T)/S(T)]$ as a function of temperature. We see that μ_N is much larger than the Hall mobility presented in Fig. 3(c). This implies that an *anomalously large* Nernst effect is observed in these materials (see discussion). Moreover, as for the Hall mobility, the Nernst mobility reaches its largest value for the optimum doping. When comparing the temperature dependence of μ_H and μ_N for the optimally doped thin films, we observe that the largest value reached by the Hall mobility at approximately 60 K corresponds roughly to the sharp downward curvature of the Nernst mobility at 75 K. Since the Nernst coefficient remains positive for all oxygen contents, the sign of μ_N is entirely determined by the sign of the thermoelectric power.

C. Mobility ratio

We calculate the ratio of the Nernst mobility to the Hall mobility in Fig. 5. In general, this ratio is increasing linearly with temperature, except for RED which crosses zero Hall mobility at the temperature shown by the dashed line, resulting in a singularity. These data indicate that μ_N is always at least 10 times larger than μ_H, except at low temperatures. Looking at the general progression with doping, this ratio is decreasing in magnitude as the oxygen content is increased (toward the antiferromagnetic-insulator phase).

IV. DISCUSSION

In the previous sections, we reported a set of transport properties from which we now expect to extract additional

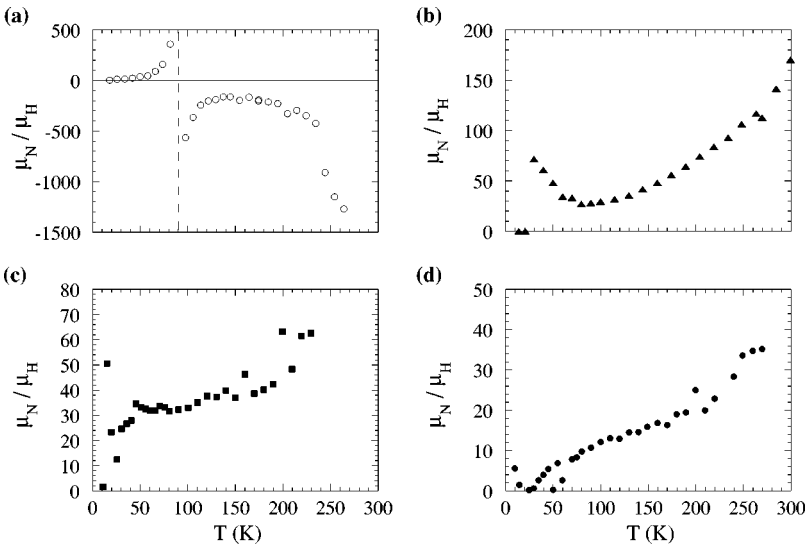

FIG. 5. The ratio of the Nernst and the Hall mobilities as a function of temperature. From top to bottom: (a) RED, (b) OPT, (c) OX, (d) SOX.

information to elucidate the scattering mechanisms of the electron-doped cuprates. In the following, we explore the applicability of several models used to explain the transport properties of the hole-doped cuprates.

A. Single-carrier transport

Simple single-carrier Boltzmann transport with a spherical Fermi surface and isotropic relaxation time[24] is unable to explain the large temperature dependence of the Hall coefficient in the hole and electron-doped cuprates. More elaborate models taking into account the appearance of two relaxation times in the resistivity and Hall measurements of hole-doped cuprates[25,26] have been proposed. Anderson[27] suggests that such duality of the relaxation times might be the result of spin-charge separation. The resistivity, only sensitive to the relaxation of momentum normal to the Fermi surface, arises from holon-spinon scattering, resulting in a relaxation time $\tau_{\rm tr} \propto T^{-1}$ ($\sigma_{xx} \propto \tau_{\rm tr}$). In the presence of a magnetic field, a second component influenced by spinon-spinon scattering (relaxation of momentum tangent to the Fermi surface) is involved, giving $\tau_H \propto T^{-2}$ ($\sigma_{xy} \propto \tau_{\rm tr}\tau_H$). This leads naturally to a Hall angle $\Theta_H \equiv \omega_c \tau_H \propto T^{-2}$ ($=R_H B/\rho_{ab}$), a behavior which is found in the hole-doped cuprates.[25] In Fig. 6, we show the inverse Hall angle for our samples as a function of T^2, clearly indicating that a higher T power is present for high temperatures:[28] we actually find that it is very close to T^4 (not shown here). We conclude that the spin-charge separation hypothesis cannot be applied *directly* to the high-temperature normal-state transport of electron-doped cuprates. This mechanism would need to explain why negative and positive carriers seem to coexist in the electron-doped cuprate family and the fact that Θ_H has a temperature power very close to 4.

Several authors propose that the anomalous Hall effect in the cuprates might be explained by a single-carrier model with an anisotropic mean-free path (MFP).[26,29] Taking two characteristic MFP's, say l_f and l_c ($l_f < l_c$) on different parts of the Fermi surface (f: on the flat parts, and c: on the curved parts), this model predicts that the conductivity is then proportional to the smallest MFP, $\sigma_{xx} \propto l_f$, while the off-diagonal component is the product of both, $\sigma_{xy} \propto l_f l_c$ (thus, the Hall angle is proportional to l_c alone). With the appropriate temperature dependences, it is then possible to reproduce qualitatively the Hall angle data of the hole-doped cuprates. If we try to apply this model to transport in NCCO at high temperature, then from resistivity, we find $l_f \propto 1/(T^2+C)$ where C is a constant. From the Hall angle (in Fig. 6), we deduce that the l_c should follow $l_c \propto 1/(T^4+C')$ where C' is a constant. This temperature dependence of the MFP is quite unusual at high temperatures, and an exotic scattering mechanism would be needed to explain it. Moreover, as in the spin-charge separation picture, this model is unable to reproduce directly the sign change observed in the temperature dependence of R_H (e.g., RED).

In general, the thermoelectric and thermomagnetic effects of cuprates are difficult to explain in the framework of conventional transport. It was observed in most hole-doped materials that the thermoelectric power can change sign as a function of doping. As hole doping is increasing in hole-doped cuprates, the high-temperature TEP (say, at 300 K) changes from positive to negative,[30] while in our case, it changes from negative to positive as electron doping is increased (decreasing oxygen content), suggesting a symmetry in doping. Some recent work[31] has proposed that the thermoelectric power of the underdoped hole-doped cuprates could reveal the opening of a pseudogap. The pseudogap increases the thermoelectric power giving a broad maximum just above T_c (and below a temperature T_g defined as the gap

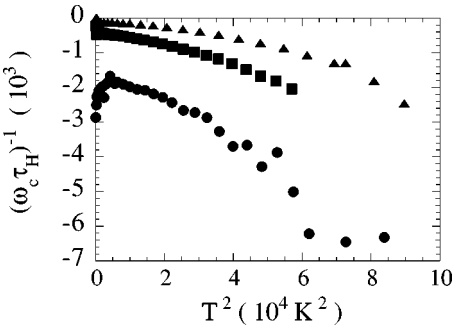

FIG. 6. Inverse Hall angle for our thin films as a function of T^2. SOX, solid circles; OX, solid squares; OPT, solid triangles. RED is not shown for clarity (see text).

temperature), the enhancement being suppressed by a relatively small amount of $3d$-metal substitution (impurities) for copper.[31] For the NCCO family, the maximum TEP is observed at fairly high temperature with respect to T_c for all the films (above 50 K). When compared with the data of Ref. 31 on YBCO, the TEP of our thin films mimics that of the Zn-doped YBCO. Because we suspect the excess oxygen to act as an impurity, and that our thermoelectric measurements also show that $S(T)$ and T_c are strongly affected by slight variation of the oxygen content, it is possible that the same mechanism leading to T_c and pseudogap suppression in Zn-doped YBCO is relevant for the electron-doped cuprates with excess oxygen. In the NCCO case, even the optimally doped (OPT) material is affected. However, to our knowledge, there has been no report of a pseudogap signature in the electron-doped cuprates.

The Nernst coefficient of our thin films is at least an order of magnitude larger than the previous reports on hole-doped cuprates.[32,33] As shown with the mobility ratio in Fig. 5, a large difference is observed between the mobilities as measured by electrical and thermal transport. In the framework of conventional Boltzmann transport theory applied to a metal with a single type of carrier and an isotropic mean free path (MFP),[34,35] the thermoelectric power and the Nernst coefficient are given, respectively, by

$$S=\pm\left(\frac{\pi^2 k_B^2 T}{3e}\right)\left[\frac{\partial \ln\sigma(E)}{\partial E}\right]_{E_F} \tag{1}$$

and

$$Q=\left(\frac{\pi^2 k_B^2 T}{3m^*}\right)\left[\frac{\partial \tau_{\rm th}(E)}{\partial E}\right]_{E_F}, \tag{2}$$

where $\tau_{\rm th}$ is defined as the relaxation time measured by *thermal* transport, m^* is the effective mass (in absolute value), and E_F is the Fermi energy. The sign in Eq. (1) is determined by the sign of the free carriers (e is a positive constant). Whereas the TEP theory has been extensively developed (see, for example, Ref. 35), the Nernst coefficient is barely mentioned in textbooks. The obvious reason is that, for most materials, the evaluation of the Nernst coefficient implies the knowledge of the detailed energy dependence of the relaxation time. Of course, the potential anisotropy of the mean free path (detail of the Fermi surface) and the combined contributions from different sources of scatterers are additional obstacles in deducing a clear (quantitative) picture for the scattering mechanisms. However, it is possible to get a rough estimate of the order of magnitude of this coefficient using Eq. (2). In general, one can assume that the energy dependence of the relaxation time follows: $\tau_{\rm th}(E)=\tau_o E^p$,[24,35] giving $[\partial\tau_{\rm th}(E)/\partial E]_{E_F}=p\,\tau_{\rm th}(E)/E_F$, and $Q\approx p\,\mu_{\rm th}$ $(\pi^2 k_B/3e)(k_BT/E_F)$. Here we have defined $\mu_{\rm th}\equiv e\tau_{\rm th}/m^*$. The value of the exponent p depends on the nature of the scatterers, but ranges from 3/2 to $-1/2$. If we substitute the Hall mobility ($\mu_H\equiv e\tau_H/m^*$) for the *thermal* mobility (in absolute value) and evaluate Q at 100 K for the optimally doped thin film, we get an absolute value of 1–4 nV/K T ($k_BT/E_F\approx 1/100$), at least one order of magnitude lower than our measured value of $\approx$80 nV/K T. Interestingly, this estimate of the Nernst coefficient approaches the measured values in the hole-doped cuprates.[32,33] This leads us to speculate that the Nernst coefficient of the hole-doped cuprates might be more conventional (it has the right order of magnitude), but it is anomalously large in the electron-doped cuprates.

This can be emphasized even more by evaluating the mobility ratio μ_N/μ_H using Eqs. (1) and (2):

$$\frac{\mu_N}{\mu_H}=\left(\frac{\tau_{\rm th}}{\tau_H}\right)\left[\frac{(\partial\ln\tau_{\rm th}/\partial E)_{E_F}}{N(E_F)/n+(\partial\ln\tau_{\rm th}/\partial E)_{E_F}}\right], \tag{3}$$

where $N(E_F)$ and n are the density of states at the Fermi level and the density of free carriers, respectively. In conventional transport, the first term in the right part of Eq. (3) is equal to 1 (in the single-carrier model). Since $\{\partial\ln\tau_{\rm th}(E)/\partial E\}_{E_F}=p/E_F$ (with p usually ranging from $-1/2$ to 3/2) and $N(E_F)/n=C/E_F$ ($C=3/2$ and 1 for a spherical three-dimensional Fermi surface and cylindrical two-dimensional Fermi surface, respectively), the second term of Eq. (3) should remain smaller than 1 in absolute value. Equation (3) could result in a large *negative* value only if the denominator is tending to zero. This would happen if $\{\partial\ln\tau_{\rm th}(E)/\partial E\}_{E_F}=-N(E_F)/n$, or $p/E_F=-N(E_F)/n$, resulting in unphysical values of p, lower than -1, for both the spherical and the cylindrical Fermi surfaces. We conclude, within the previous crude assumptions (in particular the shape of the Fermi surface), that μ_N should always be *smaller* than μ_H in magnitude (because the second term is always smaller than 1). Obviously, our observations in Fig. 5 (that $\mu_N/\mu_H>1$ for all temperature) are far from the conclusion of this simple theory. Moreover, our measurements yield a large *positive* value, inconsistent with the negative value expected if the denominator of Eq. (3) tends to zero (for negative p). Interestingly, a significant temperature dependence for the mobility ratio (as observed in Fig. 5) is only possible if $\tau_{\rm th}\neq\tau_H$.

Some authors have suggested that a van Hove singularity close to the Fermi level might influence significantly many of the normal-state properties and be at the origin of superconductivity in the cuprates.[36] This could affect strongly $N(E_F)$ and $(\partial\tau_{\rm th}/\partial E)_{E_F}$, and thus μ_N/μ_H in Eq. (3). However, at first sight, such singularities would increase $N(E_F)$, leading to a lower μ_N/μ_H. Since NCCO is not very different from the other cuprates (apart from the sign of the dominant carriers), one would expect a similar influence of the van Hove singularity on its properties. However, photoemission spectroscopy in NCCO (Ref. 37) shows a singularity many tens of meV away from E_F (further away than hole-doped cuprates[38]), thus unlikely to influence directly transport.

B. Two-carrier transport

The smooth variation of the Hall coefficient as a function of oxygen content is difficult to explain with a single-carrier model: it is even more difficult to imagine how a single type of carrier could produce a sign change of the Hall effect with temperature. In Refs. 7 and 12, it is suggested that the NCCO family could actually be described by a two-carrier model, based on a complete set of transport measurements at 60 K. For NCCO, *a positive-carrier component appears as one*

tries to increase the electron density (by reduction, as in Fig. 1). In a two-carrier system, the Hall coefficient can be expressed as

$$R_H \approx \frac{R_p\sigma_p^2 + R_n\sigma_n^2}{(\sigma_p+\sigma_n)^2} \tag{4}$$

where the R_i's and σ_i's are the Hall coefficients and the conductivities of the ith type of carriers. Then, it is easy to understand that a temperature-dependent Hall coefficient could arise if two types of charge carriers with different signs and different temperature dependences for their mobility (and relaxation time) are present. Assuming that a continuous decrease of σ_n/σ_p accompanies the oxygen depletion, R_H can then be driven from a negative value (R_n), when $\sigma_n \gg \sigma_p$, to a positive one (R_p), when $\sigma_n \ll \sigma_p$. Crusellas *et al.*[39] showed that this model can reproduce quantitatively the resistivity and the Hall coefficient, and qualitatively the magnetoresistance of $Sm_{2-x}Ce_xCuO_{4+\delta}$ single crystals.

With the same two-carrier model, the TEP can be derived as[40]

$$S(T) = \frac{S_p\sigma_p + S_n\sigma_n}{\sigma_p+\sigma_n}, \tag{5}$$

where the σ_i's and S_i's are, respectively, the conductivity and TEP of the ith carrier (the S_i's carry the sign of their corresponding charge carriers). It was previously suggested that the small variations of the oxygen content in NCCO (on the order of 0.01 to 0.04 per unit formula[41]) does not affect significantly the density of carriers, but rather modifies their mobility.[22] This effect could arise if the additional oxygen found at the apical site by neutron scattering[42] contributes to scattering. The variation of $S(T)$ with doping [in Fig. 4(a)] can be simulated in Eq. (5) if we assume (as we did above for R_H) that the temperature-dependent ratio of σ_n/σ_p is decreasing with decreasing oxygen content. This implies that the TEP is smoothly going from S_n to S_p. This variation of $S(T)$ can then be directly correlated to the variation of R_H with oxygen content. Of course, the ratio S_n/S_p might also change with doping, and makes a quantitative analysis more difficult. Our thermoelectric power measurements alone cannot confirm the simultaneous presence of two types of carriers (positive and negative) as a simple band-filling scenario could also explain the sign change.

To explain the anomalously large magnitude of the Nernst effect, we can use Sondheimer's expression[40] for the Nernst coefficient given as

$$Q(T) = \frac{Q_p\sigma_p + Q_n\sigma_n}{\sigma_p+\sigma_n} + \frac{\sigma_p\sigma_n(S_p-S_n)(\sigma_pR_p-\sigma_nR_n)}{(\sigma_p+\sigma_n)^2}, \tag{6}$$

where the Q_i's are the ith band Nernst components given by Eq. (2). The first term in Eq. (6) is behaving as the TEP in Eq. (5). As σ_n/σ_p decreases, this first term is smoothly going from Q_n to Q_p, and its magnitude should be of the same order of magnitude as the previous estimate ($Q_i \approx 1$–4 nV/K T). The second term can be responsible for the potentially large Nernst coefficient with respect to single-carrier theory. The factor $(\sigma_pR_p-\sigma_nR_n)=(\mu_p-\mu_n)$ reaches a maximum value when the mobilities are large and $\mu_p=-\mu_n$ (note that the μ_i's carry the sign of their charge). To estimate the corresponding value, let suppose that both μ_p and μ_n at low temperatures are of the order of 20–30 cm^2/V s [estimated from the result of Fig. 3(c)], that both S_p and S_n are in the range 2–5 μV/K [from Fig. 4(a)], and that $\sigma_p\sigma_n/(\sigma_p+\sigma_n)^2$ is close to 1/4 (its maximum value), then we obtain a magnitude of $Q \approx 10$–20 nV/K T . Although this estimate is still lower than the measured value, it might be easily underestimated by a factor of 5–10 if one is choosing values which are too low for the S_i's and σ_iR_i's. Nevertheless, it shows clearly how a large Nernst effect can be obtained in the two-carrier model.

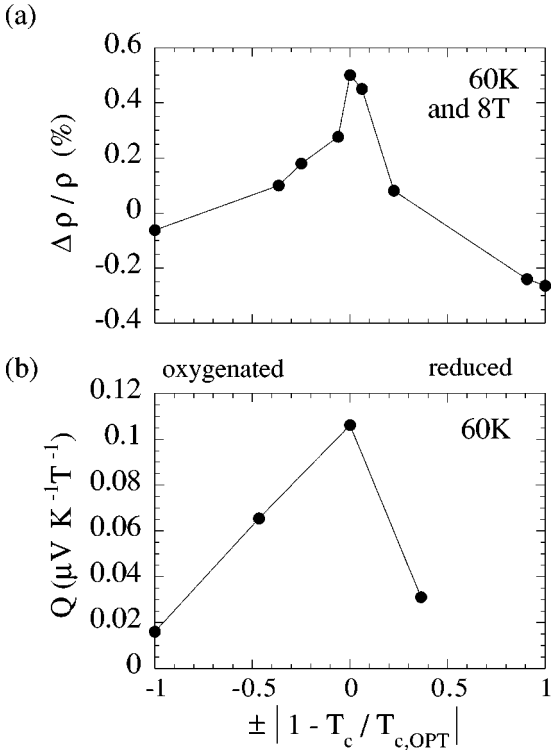

FIG. 7. Magnetoresistance (top) and Nernst coefficient (bottom) at 60 K as a function of oxygen content for several samples. Solid lines are guides to the eye.

Previously, we found that the magnetoresistance of NCCO reaches a maximum positive value for the optimal oxygen content (maximum T_c).[12] This magnetoresistance is large for the cuprate family as the hole-doped cuprates have a value at least one order of magnitude smaller.[43,44] A two-carrier model predicts the magnetoresistance to be given by[39,45]

$$\frac{\Delta\rho}{\rho} = \frac{\sigma_p\sigma_n(\sigma_pR_p-\sigma_nR_n)^2B^2}{(\sigma_p+\sigma_n)^2} \tag{7}$$

in the small Hall angle limit (appropriate for our samples). In Eq. (7), we can identify the same mobility coefficient, $(\sigma_pR_p-\sigma_nR_n)$, obtained for the Nernst coefficient in the second term of Eq. (6). This indicates that a maximum of the magnetoresistance is likely to be coincident with a maximum of the Nernst coefficient as the doping and the mobilities are gradually changed. In Fig. 7, we show the magnetoresistance from Ref. 12 and the Nernst coefficient at 60 K as a function of oxygen content and realize that both reach a maximum at

optimum doping. This is perhaps the most compelling evidence that a two-carrier model is really describing adequately the transport of the electron-doped cuprates.

V. SUMMARY

We have measured the oxygen dependence of the electrical and thermal transport properties of optimally doped ($x=0.15$) $Nd_{2-x}Ce_xCuO_{4\pm\delta}$. We report measurements of the normal-state Nernst effect in NCCO from T_c to room temperature. We show that its anomalously large magnitude cannot be explained using conventional single-carrier transport. Moreover, the doping dependence of the Nernst coefficient and the magnetoresistance are strikingly coincident. This observation, together with the temperature dependence and oxygen content variation of the Hall coefficient, is consistent with the description of the system using a two-carrier model. However, the nature of the two different carriers still remains unknown. Although a body of evidence has been gathered in this work for conventional two-carrier transport, there might still be possible avenues for nonconventional theories. The challenge would then be to explain in detail the apparent contribution of two carriers in the transport properties of electron-doped cuprates, the systematic variations as a function of cerium and oxygen doping, and eventually the appearance of superconductivity.

ACKNOWLEDGMENTS

The authors acknowledge the fruitful discussions with Eric Maiser (Karlsruhe), T.H. Geballe (Stanford), and B. Moyzhes. We also thank J.L. Peng and Z.Y. Li for target preparation. This work was supported by the NSF Condensed Matter Physics Division through Grant No. DMR 9510475.

*Also at BDM International Inc., 5109 Leesburg Pike, Suite 100, Six Skyline Place, Falls Church, VA 22041.

†Also at Goldman, Sachs & Co., 85 Broad Street, New York, NY 10004.

‡Also at Seagate Technology Inc., 7801 Computer Ave., Minneapolis, MN 55435.

[1] S. J. Hagen, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. B **43**, 13 606 (1991).

[2] M. B. Maple, MRS Bull. **15**, 60 (1990).

[3] J. L. Peng, E. Maiser, T. Venkatesan, R. L. Greene, and G. Czjzek, Phys. Rev. B **55**, R6145 (1997).

[4] M. Brinkmann *et al.* [Phys. Rev. Lett. **74**, 4927 (1995)] suggest that superconductivity can be achieved in $Pr_{2-x}Ce_xCuO_{4\pm\delta}$ single crystals for x as low as 0.05 following a special annealing process: this has yet to be confirmed by further investigations.

[5] H. Y. Huang, B. Batlogg, H. Takagi, H. L. Kwo, R. J. Cava, J. J. Krajewski, and W. F. Peck, Jr., Phys. Rev. Lett. **72**, 2636 (1994).

[6] N. P. Ong, in *Physical Properties of High-Temperature Superconductors II*, edited by D.M. Ginsberg (World Scientific, Singapore, 1990), p. 459.

[7] Y. Iye, in *Physical Properties of High-Temperature Superconductors III*, edited by D.M. Ginsberg (World Scientific, Singapore, 1991), p. 285, and references therein.

[8] J. L. Cohn, M. S. Osofsky, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. B **46**, 12 053 (1992).

[9] S. J. Hagen, Z. Z. Wang, and N. P. Ong, Phys. Rev. B **40**, 9389 (1989).

[10] M. F. Crommie and A. Zettl, Phys. Rev. B **41**, 10 978 (1990).

[11] D. H. Wu, J. Mao, S. N. Mao, J. L. Peng, X. X. Xi, T. Venkatesan, R. L. Greene, and S. M. Anlage, Phys. Rev. Lett. **70**, 85 (1993).

[12] 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).

[13] W. Jiang, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. B **47**, 8151 (1993).

[14] J. E. Hirsch and F. Marsiglio, Physica C **162-164**, 591 (1989).

[15] M. E. López-Morales, R. J. Savoy, and P. M. Grant, Solid State Commun. **71**, 1079 (1989); S. I. Lee, Y. H. Jeong, K. H. Han, Z. S. Lim, Y. S. Song, and Y. W. Park, Phys. Rev. B **41**, 2623 (1990); X.-Q. Xu, S. J. Hagen, W. Jiang, J. L. Peng, Z. Y. Li, and R. L. Greene, *ibid.* **45**, 7356 (1992).

[16] P. Ghigna, G. Spinolo, M. Scavini, G. Chiodelli, G. Flor, and A. V. Chadwick, Physica C **268**, 150 (1996); G. S. Okram, B. D. Padalia, O. Prakash, S. K. Agarwal, and A. V. Narlikar, *ibid.* **277**, 19 (1997).

[17] A. R. Drews, M. S. Osofsky, H. A. Hoff, J. L. Peng, Z. Y. Li, R. L. Greene, and T. A. Vanderah, Physica C **200**, 122 (1992).

[18] P. Fournier, E. Maiser, and R. L. Greene (unpublished).

[19] S. N. Mao, X. X. Xi, S. Bhattacharya, Q. Li, T. Venkatesan, J. L. Peng, R. L. Greene, J. Mao, D. H. Wu, and S. M. Anlage, Appl. Phys. Lett. **61**, 2356 (1992); S. N. Mao, W. Jiang, X. X. Xi, Q. Li, J. L. Peng, R. L. Greene, T. Venkatesan, D. Prasad Beesabathina, L. Salamanca-Riba, and X. D. Wu, *ibid.* **66**, 2137 (1995).

[20] In previous papers, we believed that a peak at $2\Theta=35°$, was indicative of a CeNdO secondary phase. However, in our recent work with PrCeCuO thin films [E. Maiser *et al.* (unpublished)], $Pr_2CuO_{4+\delta}$ thin films also showed the same peak, clearly indicating that it is a (110) signature (and not a CePrO phase).

[21] W. Jiang, X. Q. Xu, S. J. Hagen, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. B **48**, 657 (1993).

[22] M. Brinkmann, T. Rex, M. Stief, H. Bach, and K. Westerholt, Physica C **269**, 76 (1996).

[23] O. G. Singh, C. S. Harendranath, O. Prakash, and B. D. Padalia, Solid State Commun. **100**, 721 (1996).

[24] J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960).

[25] T. R. Chien, Z. Z. Wang, and N. P. Ong, Phys. Rev. Lett. **67**, 2088 (1991); J. M. Harris, Y. F. Yan, and N. P. Ong, Phys. Rev. B **46**, 14 293 (1992); A. Carrington, A. P. Mackenzie, C. T. Lin, and J. R. Cooper, Phys. Rev. Lett. **69**, 2855 (1992).

[26] C. Kendziora, D. Mandrus, L. Mihaly, and L. Forro, Phys. Rev. B **46**, 14 297 (1992); N. E. Hussey, J. R. Cooper, J. M. Wheatley, I. R. Fisher, A. Carrington, A. P. Mackenzie, C. T. Lin, and O. Milat, Phys. Rev. Lett. **76**, 122 (1996).

[27] P. W. Anderson, Phys. Rev. Lett. **67**, 2092 (1991).

[28] In this figure, we have omitted sample RED as its R_H crosses zero, which gives divergences of the inverse Hall angle. Moreover, we ignore the structure for $T\leqslant 100$ K which is related to

the maximum of the Hall mobility and the localizationlike upturn in resistivity.

[29] B. P. Stojkovic and D. Pines, Phys. Rev. Lett. **76**, 811 (1996).

[30] S. D. Obertelli, J. R. Cooper, and J. L. Tallon, Phys. Rev. B **46**, 14 928 (1992).

[31] J. L. Tallon, J. R. Cooper, P. S. I. P. N. de Silva, G. V. M. Williams, and J. W. Loram, Phys. Rev. Lett. **75**, 4114 (1995).

[32] J. A. Clayhold, Phys. Rev. B **54**, 6103 (1996).

[33] N. V. Ageev, V. E. Gasumyunts, and V. I. Kaidanov, Tech. Phys. Lett. **20**, 831 (1994).

[34] F. J. Blatt, *Physics of Electronic Conduction in Solids* (McGraw Hill, New York, 1968).

[35] R. D. Barnard, *Thermoelectricity in Metals and Alloys* (Taylor & Francis, London, 1972).

[36] D. M. Newns, P. C. Pattnaik, and C. C. Tsuei, Phys. Rev. B **43**, 3075 (1991), and references therein.

[37] D. M. King, Z. -X. Shen, D. S. Dessau, B. O. Wells, W. E. Spicer, A. J. Arko, D. S. Marshall, J. DiCarlo, A. G. Loeser, C. H. Park, E. R. Ratner, J. L. Peng, Z. Y. Li, and R. L. Greene, Phys. Rev. Lett. **70**, 3159 (1993); R. O. Anderson, R. Claessen, J. W. Alen, C. G. Olson, C. Janowitz, L. Z. Liu, J.-H. Park, M. B. Maple, Y. Dalichaouch, M. C. de Aldrade, R. F. Jardim, E. A. Early, S.-J. Oh, and W. P. Ellis, *ibid.* **70**, 3163 (1993).

[38] D. S. Dessau, Z.-X. Shen, D. M. King, D. S. Marshall, L. W. Lombardo, P. H. Dickinson, A. G. Loeser, J. DiCarlo, C.-H. Park, A. Kapitulnik, and W. E. Spicer, Phys. Rev. Lett. **71**, 2781 (1993).

[39] M. A. Crusellas, J. Fontcuberta, S. Pinol, M. Cagigal, and J. L. Vicent, Physica C **210**, 221 (1993).

[40] E. H. Sondheimer, Proc. R. Soc. London, Ser. A **193**, 484 (1948).

[41] E. Moran, A. I. Nazzal, T. C. Huang, and J. B. Torrance, Physica C **160**, 30 (1989); J. L. Peng, Y. Z. Li, and R. L. Greene, in *High Temperature Superconductors: Fundamental Properties and Novel Materials Processing*, edited by D. K. Christen *et al.*, MRS Symposia Proceedings No. 169 (Materials Research Society, Pittsburgh, 1990), p. 173.

[42] A. J. Schultz, J. D. Jorgensen, J. L. Peng, and R. L. Greene, Phys. Rev. B **53**, 5157 (1996); P. G. Radaelli, J. D. Jorgensen, A. J. Schultz, J. L. Peng, and R. L. Greene, *ibid.* **49**, 15 322 (1994).

[43] J. M. Harris, Y. F. Yan, P. Matl, N. P. Ong, P. W. Anderson, T. Kimura, and K. Kitazawa, Phys. Rev. Lett. **75**, 1391 (1995).

[44] T. Kimura, S. Miyasaka, H. Takagi, K. Tamasaku, H. Eisaki, S. Uchida, K. Kitazawa, M. Hiroi, M. Sera, and N. Kobayashi, Phys. Rev. B **53**, 8733 (1996).

[45] W. Jones and N. H. March, *Theoretical Solid State Physics, Volume 2: Non-Equilibrium and Disorder* (Dover, New York, 1973), Chap. 6.