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Transport and localization in $Nd_{2-x}Ce_xCuO_{4-y}$ crystals at low doping

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We report in-plane transport properties of $Nd_{2-x}Ce_xCuO_{4-y}$ crystals prepared in low-doped ($x \sim 0.01$–0.11), nonsuperconducting compositions. The large negative Hall coefficient R_H and thermopower S in these samples confirm the presence of a small density of n-type carriers. At low T we find a highly anisotropic negative magnetoresistance which, together with the behavior of ρ, S, and R_H, demonstrates two-dimensional weak localization of these carriers occurring as Ce content decreases. Analysis of the magnetoresistance data reveals an anomalously weak T dependence of the electron inelastic-scattering rate $1/\tau_i$.

Two key issues in the study of the superconducting cuprates are the nature of the two-dimensional (2D) electronic state in the CuO_2 layers and the origin of the metallic-to-insulating transition that occurs at low carrier concentration. The low-temperature transport properties, including resistivity, Hall effect, and thermopower, offer valuable probes of the electronic state and will exhibit different behavior if the transition is due to the presence of an energy gap or the onset of localization. Magnetoresistance (MR) is of special interest, since MR measurements on weakly localized systems can reveal the characteristic elastic and inelastic electron-scattering rates and their temperature behavior, thus providing valuable insight into the electronic state at low T.

Recent studies[1–3] on nonsuperconducting $Nd_{2-x}Ce_xCuO_{4-y}$, $Bi_2Sr_2CuO_6$, and $La_{2-x}Sr_xCuO_4$ found unexpected behavior in the MR of these cuprates. While a 2D weakly localized electronic system in the CuO_2 planes would be expected to show a highly anisotropic MR, Preyer *et al.*[1] reported a negative MR in crystals of $La_{2-x}Sr_xCuO_4$ at $T < 100$ K that was independent of the angle between the field and the planes. In $Nd_{2-x}Ce_xCuO_{4-y}$ films, Tanda, Honma, and Nakayama found a negative MR that was reduced by only $\sim 50\%$ as the field was rotated into the crystal *ab* plane. These puzzling results suggest that 2D weak localization offers an incomplete description of the transition to the insulating state and that spin-dependent scattering may influence transport at low carrier concentration n. In addition, although MR is a valuable probe of the electronic state there has been no detailed study of MR in an n-type cuprate to examine the electronic scattering properties.

We have investigated transport in $Nd_{2-x}Ce_xCuO_{4-y}$ more fully by studying thermopower, resistivity, Hall effect, and magnetoresistance of crystals prepared in the nonsuperconducting regime $x < 0.1$. Our results demonstrate quite clearly that the disappearance of superconductivity at low n is accompanied by the onset of 2D weak localization. Moreover, from our MR data we find evidence for an unusual temperature dependence in the inelastic-scattering rate at low T.

We prepared large single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$ over a range of dopings $0.01 \le x \le 0.15$ from a CuO flux; details of the growth and characterization of these samples appear elsewhere.[4,5] Figure 1 shows the in-plane resistivity ρ, Hall effect R_H, and thermopower S of a $Nd_{2-x}Ce_xCuO_{4-y}$ crystal with $x \approx 0.025$. As reported earlier,[5] R_H in the low-doped (and superconducting) crystals is negative; both R_H and ρ decrease in magnitude as the Ce content x increases. From the measured R_H we estimate the low-T carrier density (in a one-band model) as $n \sim 6\times10^{19}$–$2.5\times10^{20}/cm^3$ or ~ 0.01–0.04 carriers per cell, as expected from the Ce content. The resistivity is metallic for $T > 100$ K, but shows a weak upturn at low T. Crystals with $0.01 \le x \le 0.11$ show similar behavior in ρ.

The thermopower of the low-x crystals is quite similar to S at low carrier density in the Bi-Sr-Ca-Cu-O, $La_{2-x}Sr_xCuO_4$, and $YBa_2Cu_3O_7$ systems.[6] Above 100 K S is only weakly T dependent and approaches a constant value $\sim -k_B/e$, as expected[7] for a metal with a small number of n-type carriers at high temperatures $T > \varepsilon_F/k_B$ $[\approx \pi\hbar^2 n_{2D}/(2mk_B) \sim 55$ K]. Below 40 K S shows the linear behavior expected for a metal at low T. The vanishing of the thermopower as $T \rightarrow 0$ is evidence that the upturn in the resistivity is due to localization, rather than the appearance of an energy gap at the Fermi level.[7] The appearance of an energy gap would require thermal activation of charge carriers at low T, causing divergences in

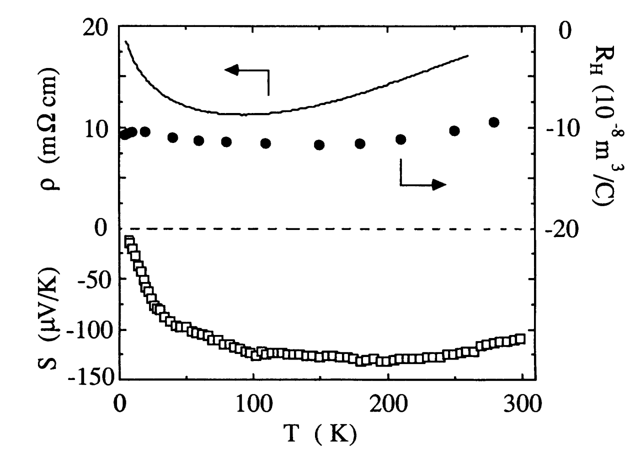

FIG. 1. Resistivity ρ, thermopower S, and Hall coefficient R_H vs T for a crystal with $x \approx 0.025$.

S and R_H at low T. Such divergences are not seen in our low-x crystals. Our MR measurements (below) demonstrate clearly that 2D weak localization is instead occurring at low T.

The resistivity ρ in the low-x crystals increases at low T with a $\ln T$ dependence, as reported for thin films.[2] Logarithmic increase of ρ is not expected in a system with an energy gap or where hopping conduction dominates. However, the theories of weak localization and Coulomb interactions both predict logarithmic corrections to the low-T conductivity $\sigma \equiv 1/\rho$, giving[8]

$$\sigma(T) = \sigma_0 + (e^2/\pi h)[(1-F) + \alpha p]\ln T/T_0 , \qquad (1)$$

where F and $\alpha \le 1$ are constants and p describes the inelastic-scattering rate through $1/\tau_i \sim T^p$. The term in $1-F$ results from Coulomb interactions, while the αp term comes from localization. Conductance measurements alone cannot distinguish these contributions, but because the two phenomena produce MR with different field and angular dependence MR measurements can be used to separate them: 2D localization produces a negative MR for fields $H \perp ab$ and none for $H \parallel ab$, while the interaction MR is isotropic in field direction.

We mounted the crystals onto a rotatable stage in a superconducting solenoid so that the angle between the field and crystal c axis could be changed continuously and the anisotropy of the MR obtained. With a Hall sensor fixed to this stage we could easily orient the stage with respect to the field to an accuracy $< 0.1°$. Using carbon-glass resistor thermometry (compensated for the small magnetoresistance of the sensor at high fields and low temperatures) we obtained temperature stability better than ± 20 mK as the field varied. Figure 2 shows the MR of a crystal with $x \approx 0.01$ at three temperatures for field orientations $H \perp ab$ and $H \parallel ab$. The MR is large and negative for $H \perp ab$ ($\sim -9\%$ at 5 K and 65 kG) but positive and much smaller for $H \parallel ab$ ($< 0.5\%$ at 5 K). We obtained the anisotropic magnetoconductance from the difference $\Delta\sigma(H) = \sigma(H \perp ab) - \sigma(H \parallel ab)$. Since the interaction and localization MR are additive to leading order, this procedure isolates the localization MR.

Weak localization is an interference phenomenon occurring in disordered conductors at low T as the carrier inelastic mean free path l_i becomes large. An electron's amplitude for diffusing around a closed path interferes constructively with that for the time-reversed path, so the total amplitude at the starting point is enhanced and the conductivity is suppressed. Applying a magnetic field perpendicular to the plane of motion destroys this constructive interference once the area l_i^2 encloses roughly one flux quantum, i.e., at fields $H_\perp \sim hc/l_i^2 e$. In a two-dimensional metal (i.e., $l_i \gg d \equiv$ the conducting layer thickness), this causes negative MR for fields perpendicular to the plane but no effect for fields parallel to the plane. This anisotropic MR has been demonstrated, in quantitative agreement with theory, in a large number of 2D systems, including thin elemental and alloy films and silicon field-effect transistor devices.[8-10]

The magnetoconductance of a weakly localized 2D system was calculated by Altshuler *et al.*[11] and extended to the case of spin-orbit and impurity-spin scattering by Hikami, Larkin, and Nagaoka[12] and Maekawa and Fukuyama.[12] The 2D conductance $\Delta\sigma_{2D}(H) \equiv \Delta\sigma(H) \times d$ in a perpendicular field H is then[12]

$$\Delta\sigma_{2D}(H) = \alpha e^2/(\pi h)[\psi(\tfrac{1}{2} + a_1/H) - \psi(\tfrac{1}{2} + a_2/H) - \ln(a_1/a_2)] , \qquad (2)$$

where $\psi(x)$ is the digamma function, $a_1 = \hbar c/4el_i^2$, and $a_2 = \hbar c/2el_e^2$. A value $\alpha = 1$ is expected, although strong spin-orbit or magnetic scattering (relative to inelastic scattering) could produce $\alpha < 1$. Spin-orbit scattering produces an *antilocalization* effect,[13] or positive low-field MR; however, since we find only negative MR for $H \perp ab$ (above 1.6 K) there is no evidence for such a contribution in this system. Treating the CuO_2 planes as the 2D layers (i.e., $\sigma_{2D} = \sigma d$, $d \approx 6$ Å) we fit the measured $\Delta\sigma$ by (2) and obtained the inelastic and elastic mean free paths l_i and l_e as functions of temperature. Above ~ 40 K the MR for both field orientations disappears. Figure 3 shows

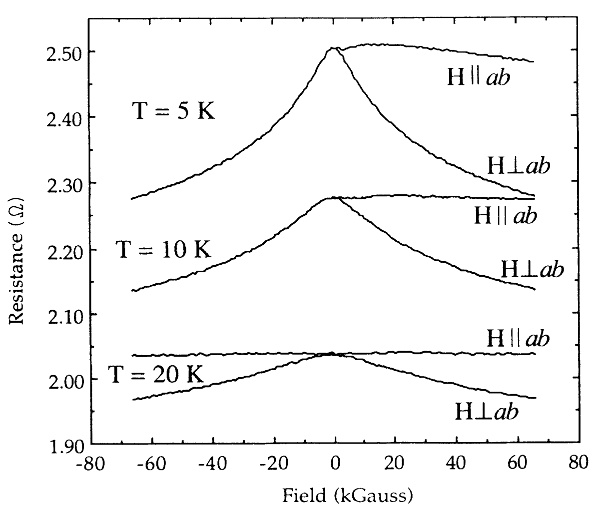

FIG. 2. Resistance vs field for an $x \approx 0.01$ crystal in fields $H \parallel ab$ and $H \perp ab$.

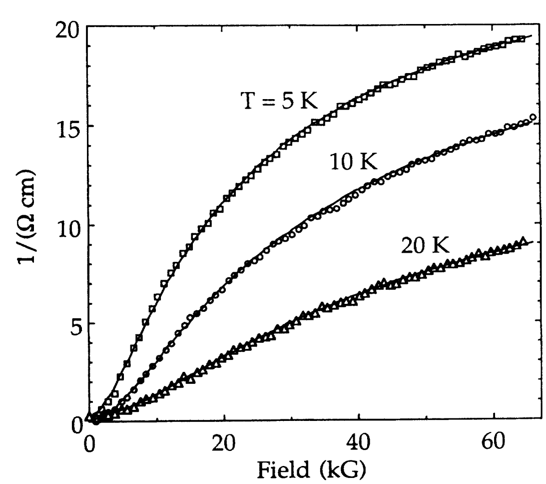

FIG. 3. Magnetoconductance vs field of an $x \approx 0.01$ crystal, with fits by Eq. (2).

typical fits by (2) for the crystal in Fig. 2; Fig. 4 shows the l_i values obtained from the fits. We find excellent agreement with (2) for $1.6 \le T \le 20$ K and $H \le 70$ kG with $l_e \sim 150$ Å and $l_i \sim 200$–300 Å. Below 10 K, l_i shows a weak power-law behavior $l_i^2 = D\tau_i \sim T^{-0.4}$ (D is the diffusion constant). For the sample in Figs. 2-4 we have $D \approx 0.7$ cm^2/s, so that $\tau_i \sim 10^{-11}$ s and $\hbar/\tau_i \sim 0.2k_BT$ at $T=5$ K. At higher temperatures the scattering rate must cross over from $T^{0.4}$ behavior to the roughly quadratic dependence $\hbar/\tau \sim k_BT^2/(75\text{ K})$ indicated by the high-T resistivity data; this evidently occurs at $T \sim 10$–20 K.

An electronic system becomes 2D with respect to electron-electron (e-e) interactions when[8] $D/d^2 \gg k_BT/\hbar$. Since $D\hbar/k_BTd^2 \sim 300$ at 5 K we compare the results for τ_i with the prediction of Abrahams *et al.*[14] for the e-e inelastic-scattering rate in a disordered 2D metal at low T. Abrahams *et al.* obtained $1/\tau_i = k_BT/(2Dm) \times \ln(T_1/T)$, where $k_BT_1 \sim 1$ Ry (13.6 eV). This gives an inelastic time $\tau_i \sim 2\times10^{-13}$ s at 5 K, which is ~ 500 times shorter than the observed $\tau_i \sim 10^{-11}$ s. In addition, the $T^{0.4}$ dependence we find in the rate $1/\tau_i$ differs significantly from the linear-T prediction. Thus we find that e-e scattering predicts too large a relaxation rate (and with too strong a T dependence) to account for the data.

While it is unclear what causes this scattering, we note a similar result in the MR data of Jing *et al.* for $Bi_2Sr_2CuO_6$, where $1/\tau_i$ shows $T^{0.33}$ behavior. This raises the interesting possibility that an unusual inelastic-scattering process with weak T dependence occurs at low-T in the conducting planes of the cuprates. This would then support transport and optical evidence (from higher T) for anomalous scattering rates in the cuprates. An important difference between the two cuprates, however, appears to be the magnitude of the scattering rate. For our typical inelastic times $\tau_i \sim 10^{-11}$ s (at 5 K) we obtain $\hbar/\tau_i \sim 0.2k_BT$, indicating that the energy levels of the charge carriers in $Nd_{2-x}Ce_xCuO_4$ are generally narrower than k_BT. However, Jing *et al.* find $\hbar/\tau_i \sim 80k_BT$ in $Bi_2Sr_2CuO_6$, indicating that inelastic scattering in this material broadens the energy states considerably relative to k_BT. From this perspective the inelastic-scattering rate in $Nd_{2-x}Ce_xCuO_4$ is more comparable to that in conventional metals [cf. $\tau_i \sim 2\times10^{-11}$ s in Cu at 5 K (Ref. 10)] than to a hole-doped cuprate.

In the $H \parallel ab$ MR (Fig. 2) a positive kink at $H \sim 10$–15 kG becomes visible at $T \sim 5$ K. This anomaly grows sharper as T decreases but remains at nearly the same H from 5 to 1.6 K. It appears consistently in the $H \parallel ab$ MR at low T, although the magnitude and exact field value vary somewhat between samples. It is evidently a spin effect (since it occurs in the longitudinal MR), but apparently does not represent the electron correlation MR since the field scale ~ 10 kG is too low for the temperature: The Zeeman splitting of the electronic levels does not become important until $2\mu H \sim k_BT$. At 10 kG, $2\mu H/k_B \approx 1.3$ K. However, Ochiai *et al.*[15] have observed

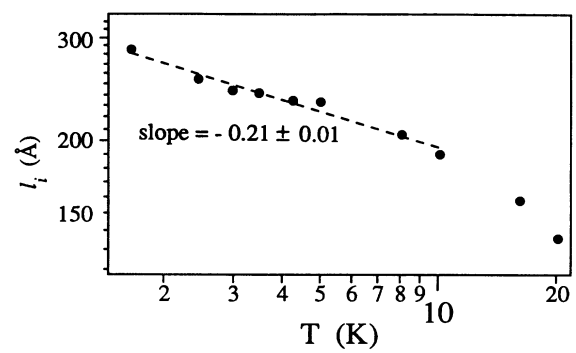

FIG. 4. Inelastic mean free path l_i vs T for an $x \approx 0.01$ crystal (log-log scale). For $T < 10$ K, $l_i \sim T^{-0.2}$.

a similar anomaly in the MR of nonsuperconducting $GdBa_2Cu_3O_7$ at the Neél temperature of the Gd ions. The kink may therefore be related to a reorientation of the Nd spins, which are believed[16] to be partially polarized at temperatures below ~ 30 K.

Disappearance of the negative MR when the field is parallel to the CuO_2 planes is very strong evidence that the low-T resistance upturn observed in $Nd_{2-x}Ce_xCuO_{4-y}$ at low x is largely due to 2D localization of charge carriers. In $Nd_{2-x}Ce_xCuO_{4-y}$ films, however, a much weaker ($\sim 50\%$) variation in the MR was reported[2] as the field was rotated into the plane. Because weak localization is so sensitive to fields perpendicular to the orbital plane of the charge carriers, this may indicate imperfect alignment of the conducting planes in the films; however, only severe misalignment ($\langle\Delta\theta\rangle \sim 30°$) could generate an isotropic MR as large as appears in the film data. A more likely origin for a negative, isotropic MR is the scattering of conduction electrons by disordered spins (which may cause isotropic MR in $La_{2-x}Sr_xCuO_4$ crystals[1]). The $Nd_{2-x}Ce_xCuO_4$ crystals studied here show long-range antiferromagnetic order below $T \sim 250$ K,[16] which would suppress spin scattering. The films, however, were prepared with Ce content x fixed near ~ 0.15 and oxygen content subsequently varied. It is not yet clear whether $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ made insulating by oxygen treatment is antiferromagnetic; if it is not, spin scattering may produce the isotropic MR.

In summary, we have studied the transport properties of single-crystal $Nd_{2-x}Ce_xCuO_4$ in the low-x range. The Hall effect and thermopower are both large and negative, indicating a small density of n-type carriers. The finite value of S at low T, along with the large negative and highly anisotropic magnetoresistance observed at low T, demonstrate that the disappearance of superconductivity is accompanied by the onset of weak localization. From the magnetoresistance data we obtain an unusual T dependence of the inelastic scattering $1/\tau_i \sim T^{0.4}$. A positive kink is observed in the longitudinal MR at moderate fields for $T < 5$ K.

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