Nature of conduction in disordered $Nd_{2-x}Ce_xCuO_{4-\delta}$ films

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We have used resistivity and Hall measurements on ion-irradiated thin films of the cuprate superconductor $Nd_{2-x}Ce_xCuO_{4-\delta}$ (NCCO) to explore the full range of transport behavior as a function of disorder in this material from superconductivity to variable-range-hopping (VRH) conduction. The cotangent of the Hall angle ($\cot\Theta_H$) in lightly irradiated samples exhibited a temperature dependence related to that of resistivity, providing support for a recent theory of Varma and Abrahams. The most highly irradiated samples exhibited $\rho(T)\propto\exp[(T_o/T)^{0.5}]$ at low temperatures, evidence of VRH in the presence of a Coulomb gap. The change of the Hall number with disorder and temperature is analyzed in the framework of possible two-carrier conduction in NCCO.

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$Nd_{2-x}Ce_xCuO_{4-\delta}$ (NCCO) has numerous unusual properties for a high-temperature cuprate superconductor, but various recent studies have elucidated the underlying similarities between its electronic properties and those of the hole-doped cuprates. It displays a similar lattice structure, similar doping phase diagram, pseudogap behavior,[1] evidence for a *d*-wave component to its order parameter,[2,3] and a similar response to point defects caused by irradiation.[4] Although NCCO exhibits unusual characteristics in magnetotransport that could be explained by two-carrier, electron-hole conductivity,[5–7] this work builds upon an earlier irradiation study in determining that NCCO shows strong parallels with the hole-doped cuprates in transport and magnetotransport.

It has been determined in many of the hole-doped cuprates near optimal doping that the cotangent of the Hall angle $\cot\Theta_H=\sigma_{xx}/\sigma_{xy}$, which is proportional to the inverse Hall mobility $\mu_H^{-1}=\rho/R_H=H\cot\Theta_H$, exhibits a quadratic temperature dependence.[8,9] In these materials, the normal-state resistivity is linear in temperature, so it has been postulated by Anderson that these two temperature dependences indicate the presence of two independent scattering mechanisms resulting from spin-charge separation.[10] The quadratic temperature dependence of $\cot\Theta_H$ in this theory is generated by spinon-spinon scattering, which follows the standard Fermi-liquid electron-electron scattering form.

A recent theory by Varma and Abrahams has posited a different cause for the quadratic temperature dependence of $\cot\Theta_H$ in the cuprates.[11] Their analysis found that $\cot\Theta_H$ should follow the square of the resistivity dependence as a general consequence of anisotropic small-angle scattering in a marginal Fermi liquid; thus, $\cot\Theta_H$ is quadratic in temperature because resistivity is linear in temperature for the hole-doped cuprates. In the case of the hole-doped cuprates, it is difficult to discriminate between the Anderson and Varma/Abrahams explanations, but for NCCO, where resistivity is not *T* linear, it should be possible. The data presented here implies that the relationship derived by Varma and Abrahams consistently describes the behavior in NCCO as well as the hole-doped cuprates.

In Ref. 4, it was shown that superconductivity is destroyed by ion irradiation in NCCO in a fashion much like in yttrium barium copper oxide (YBCO). In this present work, resistivity and Hall measurements taken on films irradiated to much higher doses cover the full range of conducting behavior from optimally superconducting to highly insulating. The variable-range-hopping insulating state accessed has the same characteristics as that seen in ion-irradiated YBCO.

Resistivity and Hall measurements were carried out on thin films of NCCO that had been patterned into Hall bars and then sequentially damaged by high-energy ions. The *c*-axis-oriented NCCO films were grown 400 nm thick using laser ablation from a target with [Ce]=0.14. A KrF (248-nm) pulsed excimer laser, focused to an intensity of approximately 1.6 J/cm^2 was used to deposit the NCCO on yttria-stabilized-zirconia (YSZ) substrates held at 820 °C in a 180-mT N_2O atmosphere. The resultant films were patterned into 100-μm-wide Hall bars using photolithography and ion milling.

Films were irradiated with 200-keV He^+ ions; these ions had enough energy to pass entirely through the NCCO and embed in the YSZ substrate, according to TRIM simulations.[12] Samples were irradiated to total doses between 5×10^{13} $ions/cm^2$ and 3.25×10^{15} $ions/cm^2$, with very low implanter current to avoid heating the films. At each damage level, $\rho(T)$ and $R_H(T)$ were measured by a standard four-point ac-lock-in technique at low frequency. Hall measurements were made at fields of ±8 T.

The wide range of irradiation doses allowed study of NCCO as a function of disorder from fully superconducting to fully insulating, covering more than seven decades of resistivity in the normal state. For low levels of damage, the resistivity curves appear parallel at temperatures above 150 K, implying they only differ by a temperature-independent scattering term, as seen in the top panel of Fig. 1. Above 150 K, the resistivity data follows Mathiessen's rule with $\rho(T)=\rho_o+A(T)$, where $A(T)$ is constant over damage levels and

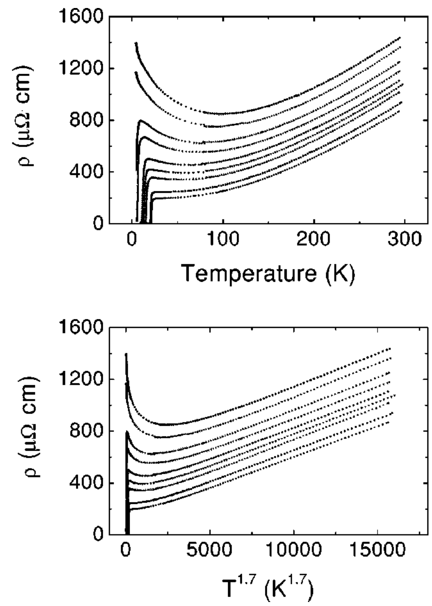

FIG. 1. (Top panel) Resistivity as function of temperature for ion-irradiated NCCO films. From bottom to top, ion doses are 0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 4.0, and 4.5×10^{14} ions/cm^2. (Bottom panel) Resistivity vs $T^{1.7}$ for the same doses appear linear at higher temperatures, implying $\tau_\rho \propto T^{-1.7}$.

ρ_o increases with irradiation dose. The resistivity curves can be well fit by $A(T)=\alpha T^{\beta}$, where $\beta=1.7$. The lower panel of Fig. 1 shows how the effective transport scattering rate appears to be proportional to $T^{1.7}$ above 150 K, whereas YBCO and the other optimally hole-doped cuprates follow a linear T dependence.

As shown in Figs. 1 and 2, the resistivity acquires a negative value for $d\rho/dT$ at low temperatures and higher irradiation doses. At the highest doses, the samples appear truly insulating at low temperatures with a temperature dependence of $\rho(T)\propto\exp[(T_o/T)^{0.5}]$. This dependence is the same as found in YBCO at high levels of irradiation[13] and indicates a disorder-induced regime of variable-range hopping. This form for the low-temperature resistivity behavior is similar to that seen in lightly doped crystalline semiconductors and high-resistivity granular metal systems. As seen in the inset to Fig. 2, the insulating NCCO films exhibit $\rho(T)\propto\exp[(T_o/T)^{0.5}]$ for temperatures below about 10 K, which shows the greatest similarity to previous data on doped, compensated crystalline semiconductors.[14,15] The fit to this temperature dependence is superior to that found by testing other possible insulating forms of $\exp[(T_o/T)^{\alpha}]$, with $\alpha=\frac{1}{4}$, $\frac{1}{3}$, or 1. This temperature dependence can be the result of variable-range-hopping (VRH) conduction in a material with a Coulomb gap, according to the theory of Efros and Shklovskii.[16] Although such a temperature dependence could also be the result of VRH in a one-dimensional system, there are no grounds for believing ion-damaged NCCO is such a system.

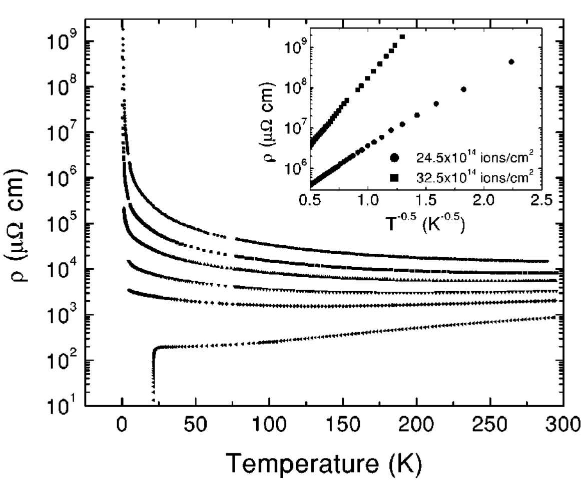

FIG. 2. High ion dose resistivity vs temperature data displays evolution to VRH insulator. From bottom to top, ion doses are 0, 6.5, 12.5, 18.5, 24.5, and 32.5×10^{14} ions/cm^2. Inset shows that $\log\rho\propto T^{-0.5}$ at low temperatures for the highest doses.

By fitting the low-temperature data of the most damaged samples to the form $\rho(T)\propto\exp[(T_o/T)^{0.5}]$, one extracts values for T_o of 6.2 K, 17.4 K, and 59.5 K, for the samples irradiated with doses of 1.85, 2.45, and 3.25 $\times10^{15}$ ions/cm^2, respectively. These values agree well with the upper temperature limit from which the $\log\rho\propto T^{-0.5}$ dependence commences for each sample, consistent with the notion that VRH conduction dominates below this temperature. According to Efros-Shklovskii, T_o is related to the localization radius a_o by $k_BT_o=2.8e^2/\kappa a_o$, where κ is the dielectric constant. It is difficult to extract a precise value for the localization length because both a_o and κ are expected to diverge around a critical carrier density according to the scaling theory of localization.[17,18] Assuming a reasonable value for κ,[19] one finds the electronic states in the material at the highest dose of irradiation are localized to a radius of $a_o\approx100$ Å.

The Hall coefficient is negative over all temperatures and irradiation levels, implying that electrons are the dominant carrier in these NCCO films. The effective Hall carrier density or Hall number ($n_H=-1/R_Hec$) rises with damage at high temperatures for doses below 1.25×10^{15} ions/cm^2, whereas at the lowest temperatures it first rises weakly and then falls with damage, as seen in the upper panel of Fig. 3. For doses below 1.25×10^{15} ions/cm^2, the phase space of this figure seems to be divided into three regimes around the point $n_H=3\times10^{21}$ cm^{-3}, dose$=3.5\times10^{14}$ ions/cm^2, and T=100 K: below 3.5×10^{14} ions/cm^2 (the dose below which the films still appear superconducting), n_H rises with dose over all temperatures displayed; at higher doses n_H falls for temperatures below T=100 K, but continues to rise for temperatures above T= 100 K. Between a dose of 1.25 $\times10^{15}$ ions/cm^2 and 1.85×10^{15} ions/cm^2, n_H collapses across the entire range of temperatures, coincident with the onset of variable-range hopping. In an earlier study on ion-irradiated YBCO (Ref. 13), it was found that n_H falls monotonically with damage at all temperatures, consistent with the notion that irradiation sequentially eliminates carriers.

A thorough study of the Hall number as a function of temperature and disorder (irradiation dose) can give a qualitative picture of how conduction evolves over the series of films measured. In YBCO n_H falls with increasing ion dose, but for NCCO n_H can rise or fall with dose. Given the ex-

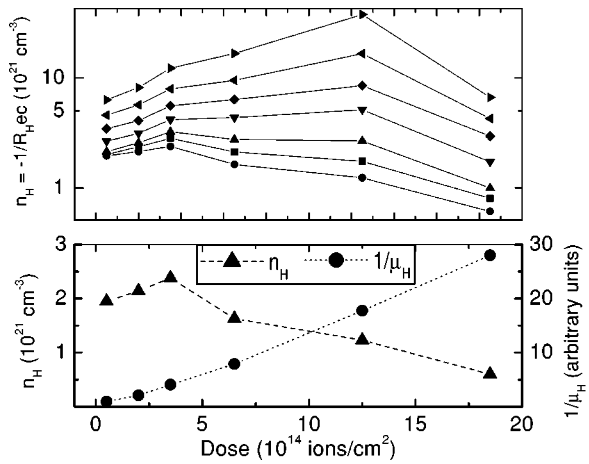

FIG. 3. (Top panel) Hall number as a function of ion dose for various temperatures. From bottom to top, temperatures are 25 K, 50 K, 75 K, 125 K, 175 K, 225 K, and 275 K. (Bottom panel) n_H and μ_H^{-1} as a function of ion dose at $T=25$ K.

isting evidence from other transport measurements for two-carrier, compensated conduction in NCCO, it is appropriate to try to interpret the change of Hall number with ion dose in NCCO in terms of relative changes in electron and hole carriers. In a two-carrier system, the total Hall number is given by $n_{H,tot}=(\rho_e+\rho_h)^2/(R_{H,e}\rho_h^2+R_{H,h}\rho_e^2)$, where $R_{H,e}$, ($R_{H,h}$), and ρ_e (ρ_h) are the Hall coefficient and resistivity for electrons (holes). In general, if carriers were lost with irradiation dose one would expect $n_{H,tot}$ to fall, but it could actually rise as both n_e and n_h fall, given the appropriate conditions. $R_{H,e}$ and $R_{H,h}$ are of opposite sign, so the denominator of $n_{H,tot}$ can approach zero when the magnitudes of $R_{H,e}\rho_h^2$ and $R_{H,h}\rho_e^2$ become equivalent, and the overall value of $n_{H,tot}$ can then diverge. To give a physical notion of when this balance condition can occur, consider the simplified case where the scattering rates and masses of the electrons and holes are approximately equal. Then $n_{H,tot}\approx(n_h+n_e)^2/(n_h-n_e)$ and $n_{H,tot}$ will diverge as the number of electron and hole carriers become nearly equal. In this case of identical carrier densities, carrier charge will segregate equally in a magnetic field, leading to a vanishing Hall voltage, which is equivalent to an infinite Hall number. A rising value for $n_{H,tot}$ with irradiation would indicate electrons were being eliminated faster than holes, equivalent to increasing compensation in this majority-electron material.

In the case of the NCCO data, it is not possible to separate effects of carrier density, scattering rate, and carrier mass, but the increase of $n_{H,tot}$ with irradiation could come from increasing balance of $R_{H,e}\rho_h^2$ and $R_{H,h}\rho_e^2$. In the top panel of Fig. 3, it is evident that the increase of n_H is most pronounced at high temperatures. At the lowest temperatures, n_H is weakly dependent on dose at low doses and then falls for all doses above 3.5×10^{14} ions/cm^2, similar to the simple result in YBCO. The lower panel of Fig. 3 shows how the Hall number (nominally proportional to carrier density) and inverse Hall mobility (nominally proportional to scattering time) vary with ion dose at the lowest temperature of 25 K. Whereas n_H rises slightly and then falls, μ_H^{-1} rises nearly

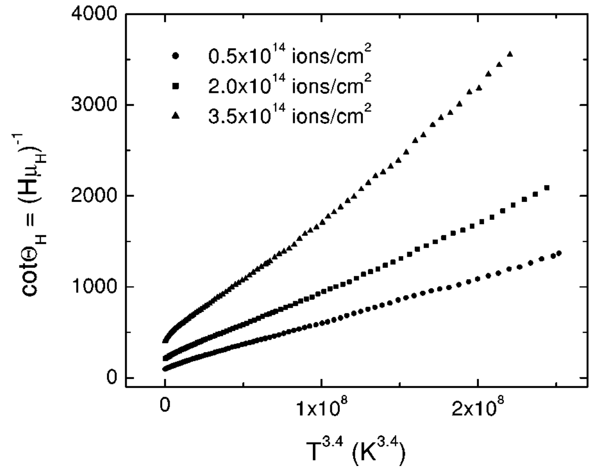

FIG. 4. Plot of $\cot\Theta_H$ vs $T^{3.4}$ for various ion doses. Data falls along straight lines implying $\tau_\mu\propto T^{-3.4}\propto\tau_\rho^2$.

linearly over all doses, implying that carrier mobility falls in a uniform fashion with increasing ion dose.

As mentioned previously, analysis of the Hall mobility brings to light an important similarity between NCCO and the hole-doped cuprates. For the hole-doped cuprates around optimal doping, the inverse Hall mobility (or $\cot\Theta_H$), closely follows a quadratic temperature dependence. For NCCO, $\cot\Theta_H$ rises much more steeply than quadratically with temperature. It does appear empirically, however, that a simple relation still exists between the temperature dependences of the resistivity and $\cot\Theta_H$. As in the hole-doped cuprates it appears that $\tau_\mu(T)\propto[\tau_\rho(T)]^2$, where τ_μ(T) is the relaxation time associated with the Hall mobility (or $\tan\Theta_H$) and $\tau_\rho(T)$ is the resistivity relaxation time [inversely proportional to ρ(T)].[20] From Fig. 1, it was found that $\rho(T)\propto T^{1.7}$, implying $\tau_\rho\propto T^{-1.7}$; Fig. 4 illustrates that $\cot\Theta_H(T)\propto T^{3.4}$, implying $\tau_\mu\propto T^{-3.4}$ is followed closely over nearly all temperatures measured. This temperature dependence persists for all damage levels up to 1.25×10^{15} ions/cm^2. Thus the relationship $\tau_\mu(T)\propto[\tau_\rho(T)]^2$ is robust in the NCCO films

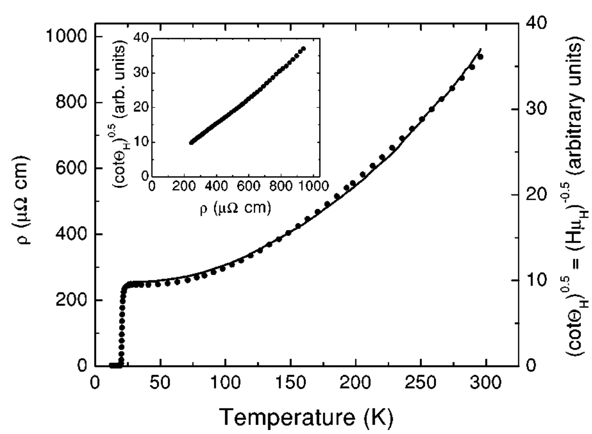

FIG. 5. Resistivity and $(\cot\Theta_H)^{0.5}$ plotted together vs temperature for the most lightly irradiated sample. The symbols track resistivity, the line tracks $(\cot\Theta_H)^{0.5}$, and the two quantities are correlated over all temperatures. The inset shows that $(\cot\Theta_H)^{0.5}$ vs resistivity defines an approximate straight line.

for all levels of disorder before VRH conduction dominates.

The recent theoretical paper [Ref. 11] by Varma and Abrahams showed that small-angle impurity scattering in a system with an anisotropic scattering rate could lead to just such a relation between the transport and mobility relaxation times. It was determined that the carriers see an effective Lorentz force created by the anisotropic nature of scattering and this produces a possibly dominant term in $\tan\Theta_H$ proportional to τ_ρ^2. For the hole-doped cuprates this would lead to $\cot\Theta_H \propto T^2$, whereas in the NCCO films studied here it would imply $\cot\Theta_H \propto T^{3.4}$. By plotting resistivity and $(\cot\Theta_H)^{0.5}$ together vs temperature (for the sample irradiated with 5×10^{13} ions/cm^2) in Fig. 5, it is seen that they are highly correlated over the entire temperature range. The inset to the figure shows that a plot of $(\cot\Theta_H)^{0.5}$ vs resistivity defines an approximate straight line, just as it does in similar data for YBCO.

An extensive view of the effect of disorder on conduction in NCCO films has been examined from 22-K superconductor to VRH insulator. For low temperatures, the inverse Hall mobility rises monotonically with damage while effective Hall carrier density is nearly constant with damage at low ion doses. At high levels of damage, μ_H^{-1} continues to rise linearly and n_H decreases, eventually collapsing at all temperatures between a dose of 1.25×10^{15} ions/cm^2 and 1.85×10^{15} ions/cm^2. As with YBCO, far in the insulating regime the material appears to conduct by variable-range hopping in the presence of a Coulomb gap. The data presented here give significant support to the recent theory of Varma and Abrahams concerning $\cot\Theta_H$ in the cuprates, which provides a physical basis for the distinctive behavior of $\cot\Theta_H$ in this class of materials. There would not appear to be a basis for the $\tau_\mu(T)\propto[\tau_\rho(T)]^2$ relationship between two nominally independent scattering times to exist in NCCO within the Anderson framework. In the cuprates, the Hall angle and resistivity do not appear to be independent parameters. Despite apparent differences, NCCO once again displays kinship with the other cuprates in its electronic properties.

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