Anomalous T^3 Inverse Hall Mobilities Observed in $Sr_{1-x}CuO_2$ and $Sr_{0.9}Nd_{0.1}CuO_2$ Infinite-Layer Thin Films

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The infinite-layer CuO_2 structure of the $Sr_xCa_yCuO_2$ and $Sr_xNd_{1-x}CuO_2$ systems are predicted to result in a simple 2D Fermi surface and a single hybridized Cu d–O p band. Unlike more complex high-T_c superconductors, the Hall mobilities in these systems should be unaffected by multiband scattering, allowing a more direct interpretation of the data. Here, we report the absence of the T^{-2} mobility commonly observed in other cuprates, suggesting that the T^{-2} dependence may originate from interband e^--e^- scattering. Finally, an anomalous T^{-3} mobility presents new challenges to our understanding.

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The Hall effect has been of great interest in the high-T_c superconductors, in part due to the unusual temperature dependence of the Hall voltage observed in the normal state, e.g., $R_H \propto 1/T$ in $YBa_2Cu_3O_7$ [1,2]. To date, no mechanism has been established conclusively as the origin of such anomalous behavior. To help in understanding such intriguing properties, intense efforts have been directed towards fabricating infinite-layer CuO_2 compounds—the basic "building blocks" of the high-T_c superconductors [3–8]. Knowledge of the transport properties in these simpler parent compounds should help provide a fundamental understanding of the physical processes that occur in all high-T_c superconducting perovskites. In this Letter, we report the first Hall measurements from epitaxial thin films of these materials, including the discovery of an anomalous temperature dependence with important theoretical implications.

The electronic band structure of tetragonal $(Ca,Sr)CuO_2$ has recently been calculated utilizing five different energy-band numerical methods [9–13]. Each approach predicts a simple 2D, nested Fermi surface originating from a single hybridized Cu $3d$–O $2p$ band for the perfect infinite-layer structure. Apart from the electron-like carriers, these structures are similar to the rounded 2D-square Fermi surface associated with the CuO_2-plane-related bands of the more complex high-T_c superconductors [14]. From these calculations, no other bands associated with the CuO_2 planes are predicted to occur within 0.4 eV of the Fermi energy E_F. In contrast to high-T_c superconductors, these materials appear to have a very low density of states (DOS) at E_F which is consistent with the higher resistivities reported in these materials [8]. If these calculations are correct, which seems likely because of their consistency with each other, physical interpretations of the Hall mobilities in these systems will not be hindered by multiband scattering processes to be discussed in more detail below.

As for many high-T_c cuprate superconductors, the band structure calculations indicate that a saddle-point singularity (SPS) occurs about 0.2 eV below E_F in the infinite-layer $(Ca,Sr)CuO_2$ phases [9–13]; this SPS is expected to occur at the Fermi surface with a strontium deficiency of $x = 0.1$ in $Sr_{1-x}CuO_2$ [13]. Great interest currently exists in characterizing high-T_c superconductors with the SPS located exactly at the Fermi energy, since it is here that superconductivity should be optimized according to BCS theory [13]. Interestingly enough, $Sr_{0.85}CuO_2$ was recently reported to have the optimized (highest) electrical conductivity over a considerable range of dopants consistent with the existence of such a SPS at E_F [8]. Therefore, Hall measurements of this phase are included in this work. Although these infinite-layer materials were studied over a wide range of divalent and trivalent cation doping, superconductivity was not observed except in the trivalent doped materials. In this class of compounds, the optimal superconducting transition temperature was found to occur in the $Sr_{0.9}Nd_{0.1}CuO_2$ phase, which was also included in the present study.

To date, two mechanisms have been proposed to account for an observed T^{-2} Hall mobility in the high-T_c superconductors. Note that it is this temperature dependence in the mobility, coupled to a linear temperature dependence of resistivity, that leads to the strongly temperature dependent Hall coefficient in $YBa_2Cu_3O_7$. The first model proposes that the T^{-2} temperature dependent mobility is due to a separation of charge (holon) and spin (spinon) quasiparticles [15], while the second suggests that it simply reflects the presence of electron-electron umklapp scattering between the electronic bands [16, 17]. In this Letter, we report the absence of this well known T^{-2} Hall mobility [18, 19] in the infinite-layer cuprate perovskites. This suggests that the T^{-2} mobility seen in most high-T_c superconductors results from the interband electron-electron scattering mechanism rather than the spinon-holon mechanism [20]. In addition, we observed the striking appearance of a clear T^{-3} Hall mobility in these compounds, suggesting the presence of a previously unobserved nonconventional scattering mechanism. This mechanism may be associated with the strong nesting of the Fermi surface predicted in recent band structure

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calculations; in any event, this apparently nonconventional scattering mechanism may be a key to understanding the pairing mechanism in the high-T_c superconductors.

To further support these claims, Hall studies also were conducted on two quenched [21] oxygen-deficient $YBa_2Cu_3O_{\sim 6.4}$ thin films. Recent band structure calculations [22] as well as experimental angle resolved photoemission spectroscopy confirmation [23] reveal that the Cu $3d$–O $2p$ plane-derived bands remain intact down to an oxygen stoichiometry of roughly $O_{\sim 6.4}$ in the Y-Ba-Cu-O system. In contrast, the metallic chain-derived bands have been shown to originate only from fully occupied chains which no longer exist in the oxygen disordered $YBa_2Cu_3O_{6.4}$ phase. Indeed, we report here that these quenched $YBa_2Cu_3O_{7-\delta}$ thin films show a conversion from the well documented T^{-2} Hall mobility (for $YBa_2Cu_3O_{7-\delta}$, $\delta < 0.55$) to a T^{-3} mobility, just before reaching the metal-insulator transition ($\delta \sim 0.6$), at which point strong localization effects dominate. This observation further supports a multiband scattering mechanism as the origin of the T^{-2} Hall mobility.

Epitaxial thin films of $Sr_{1-x}CuO_2$, $Sr_{\sim 0.9}Nd_{\sim 0.1}CuO_2$, and $YBa_2Cu_3O_{\sim 6.4}$ were grown by pulsed laser ablation [1,3]. Four-circle x-ray diffraction data, published elsewhere [3], reveal the infinite layers to be very high-quality single-crystal-like films with extremely narrow diffraction peaks, and virtually no impurity peaks present. In addition, high resolution rocking curves of the (002) reflections indicated an outstanding full width half maximum (FWHM) of ~0.14° suggesting that these films are ideal for transport studies. Briefly, simultaneous measurements were made of the resistivity, ρ, the superconducting transition temperature, T_c, and the Hall coefficient, R_H, utilizing patterned bridges [1]. Using standard dc techniques, Hall voltages were measured at a series of fixed applied fields as a function of temperature with the currents systematically reversed to eliminate thermal voltages. Afterwards, identical data-taking sequences were repeated in reversed applied fields. The final Hall coefficients were obtained by Lagrange interpolation of the two temperature sweeps followed by the subtraction defined by

$$R_H = \frac{\rho_{xy}(H) - \rho_{xy}(-H)}{2H}.$$

This process eliminated all offsets associated with the small misalignment of the Hall probes. (Hall coefficients obtained by this method are completely consistent with determinations made by holding the temperature fixed and sweeping the field from −8 to +8 T).

Figure 1 shows representative data for epitaxial thin films of $Sr_{0.85}CuO_2$, $Sr_{0.9}Nd_{0.1}CuO_2$, and quenched $YBa_2Cu_3O_{6.4}$ used in computing the inverse Hall mobilities $\cot\theta_H = E_x/E_y = (\omega\tau)^{-1}$ reported here. These inverse mobilities were then fit to a T^n form with the exponents $n = 3.0 \pm 0.1$ giving the best fit for the infinite-layer cuprates and $n = 2.0 \pm 0.1$ giving the best fit for the more complex $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.5}$ cuprates. Figure 2 illustrates these inverse Hall mobilities as a function of T^3. Except for the end-point insulating phases $SrCuO_2$ and $YBa_2Cu_3O_6$, which show strong localization effects, and $YBa_2Cu_3O_{7-\delta}$ ($\delta < 0.55$), which shows the well known $\cot\theta_H = \alpha T^{2.0\pm0.1} + \beta$ power law behavior, the remaining simpler infinite-layer materials show an anomalous $\cot\theta_H = \gamma T^{3.0\pm0.1} + \epsilon$ behavior from

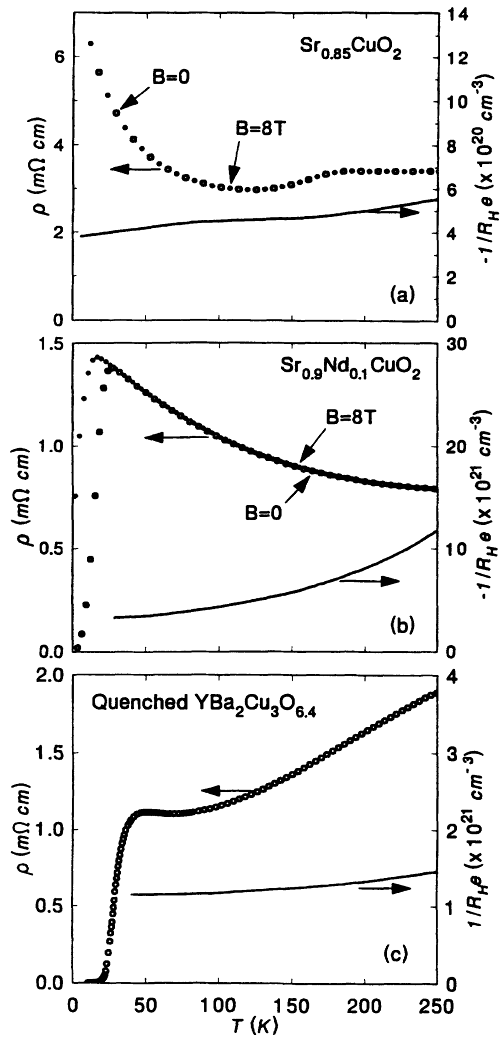

FIG. 1. Temperature dependence of the resistivity and inverse Hall coefficients for the thin-film infinite-layer compounds (a) $Sr_{0.85}CuO_2$, (b) $Sr_{0.9}Nd_{0.1}CuO_2$, and (c) oxygen-deficient $YBa_2Cu_3O_{6.4}$. The insensitivity of the resistivity to magnetic field suggests that the anomalous dip in resistivity for $Sr_{0.85}CuO_2$ is not associated with superconductivity. In contrast, both $Sr_{0.9}Nd_{0.1}CuO_2$, and $YBa_2Cu_3O_{6.4}$ are superconducting. The $YBa_2Cu_3O_{6.4}$ sample was quenched from 200 °C and cooled to below 250 K in less than 20 min after growth to minimize the effects of oxygen vacancy ordering observed in earlier studies. The Hall coefficient is negative for both infinite-layer materials and positive for $YBa_2Cu_3O_{6.4}$.

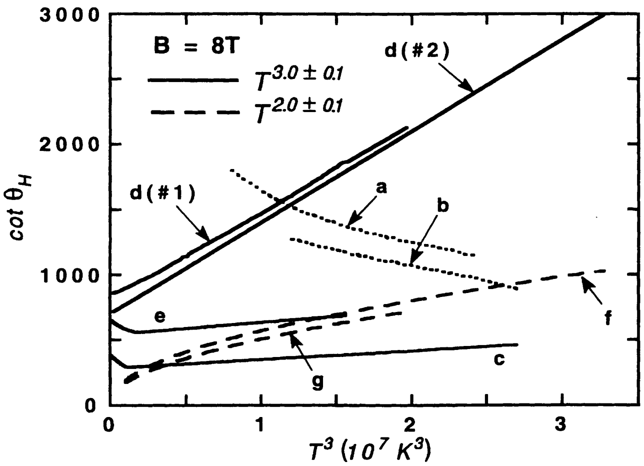

FIG. 2. Inverse Hall mobilities as a function of T^3 for the various thin films studied. The dotted curves represent the insulating phases curve a, $SrCuO_2$ and curve b, $YBa_2Cu_3O_6$ which show strong localization at lower temperatures. The more conductive phases (solid curves) curve c, $Sr_{0.85}CuO_2$, curve d, $Sr_{0.9}Nd_{0.1}CuO_2$ (2 films shown), and curve e, $YBa_2Cu_3O_{6.4}$ show an anomalous $T^{-3.0\pm0.1}$ Hall mobility above ~100 K, suggesting the presence of a nonconventional scattering mechanism in these simpler cuprates. In contrast, the high-T_c superconductors having complex electronic band structures generally show a $T^{-2.0\pm0.1}$ Hall mobility (short dashed curves), indicative that interband electron-electron scattering may be responsible for this temperature dependence. Examples include the ortho-I phase curve f, $YBa_2Cu_3O_7$ and ortho-II phase curve g, $YBa_2Cu_3O_{6.5}$. These T^2 inverse Hall angles, which clearly show curvature when plotted on the T^3 axis, may obscure the presence of a T^{-3} Hall mobility in multiband superconductors.

about 100 to 320 K (the highest temperature to which data were taken from $Sr_{0.9}Nd_{0.1}CuO_2$). In an attempt to better understand these results, the possible implications of Fermi surface nesting on Hall effect will be described in more detail below.

Several mechanisms have been proposed by others to account for temperature dependent Hall coefficients in metals [15,24,25]. However, all of these models either assume a separation of charge and spin quasiparticles [15], require the presence of both holes and electrons [24] (termed "compensation"), require multiband scattering processes [24], or assume an anisotropic basal plane scattering mechanism [25]. Therefore, interpretations of the Hall coefficient R_H are at best difficult in multiband metals. This fact motivated this study of these plausible single-band infinite-layer CuO_2 parent compounds. The important conclusion that follows from the lack of the T^{-2} mobility in these infinite-layer compounds is that electron-electron scattering between the different electronic bands may be responsible for the well known T^{-2} power law [16] in the multiband superconductors.

Although the infinite-layer cuprates are believed to be single-band materials, band structure calculations demonstrate strong nesting [13] similar to the nesting in many other high-T_c superconductors [14]. To see how nesting might influence the Hall effect in 2D metals, recall that Ong [25] showed the Hall coefficient can be represented in mean free path space by

$$R_H \propto A_l / l_{\mathrm{av}}^2 ,$$

where A_l is the "Stokes" area swept out by the scattering path length vector $\boldsymbol{l}_{\mathbf{k}}$ as $\mathbf{k}$ circumscribes the Fermi surface (FS) and l_{av} is the average mean free path. If nesting occurs, the nested states may account for only a small contribution to the temperature dependence of the Hall coefficient R_H, since in the isotropic-τ approximation, these states have the same scattering path length vector $\boldsymbol{l}_{\mathbf{k}}$. In contrast, these states can represent a significant proportion of the total longitudinal conductivity due to their large contribution to the partial electronic density of states. On the other hand, the remaining states, i.e., rounded corners of the Fermi surface, contributes more significantly to the "Stokes" area A_l thereby allowing these states to dominate more heavily in the temperature dependence of the Hall coefficient. As a consequence, the Hall angle and longitudinal conductivity are not equally sensitive to the same scattering mechanisms that occur along a nested Fermi surface. Recently, Carrington *et al.* discovered that the Anderson formula, $\cot\theta_H = \alpha T^2 + \beta$, was independent of cobalt doping in the $YBa_2(Cu_{1-x}Co_x)_3O_{7-\delta}$ system, although cobalt doping gave rise to an unusually downward curved longitudinal resistivity [17]. This finding led Carrington *et al.* to suggest electron-electron umklapp scattering as the mechanism dominating the temperature dependence of the Hall angle in $YBa_2Cu_3O_7$ while another scattering mechanism dominated the longitudinal resistivity [17]. In this work, the T^{-3} Hall mobility was found to occur in various infinite-layer cuprate perovskites, although the resistivities varied greatly from sample to sample (see Figs. 1 and 2); such observations may also be resolved in terms of such Fermi surface nesting.

In addition to the lack of the T^{-2} Hall mobility in the infinite-layer materials, the interesting observation of the T^{-3} Hall mobility suggests the presence of a nonconventional scattering mechanism that may be obscured by the T^{-2} Hall mobility in the other high-T_c materials. To the best of our knowledge, this is the first reported observation of such anomalous Hall mobilities; they cannot be explained by any presently known mechanism, including those associated with inhomogeneities [1], impurity scattering, electron-phonon scattering, and electron-electron scattering [26]. In addition, since both quenched $YBa_2Cu_3O_{6.4}$ and $Sr_{0.9}Nd_{0.1}CuO_2$ clearly show superconductivity, this nonconventional scattering mechanism, possibly associated with magnetic ordering effects, may provide a clue to the pairing mechanism in the high-T_c superconductors.

In summary, we studied the longitudinal and Hall conductivities in epitaxial thin film samples of $Sr_{1-x}CuO_2$

and $Sr_{\sim 0.9}Nd_{\sim 0.1}CuO_2$ in order to develop some new physical insight into the normal state properties of superconducting cuprate perovskites. Since the infinite-layer materials are believed to possess only the simple 2D Fermi surface that is associated with the hybridized $Cu\,3d-O\,2p$ band (as in the CuO_2 plane-related bands of the more complex high-T_c superconductors), these materials are well suited for Hall effect studies. Unlike the more complex cuprate perovskites, the T^{-2} Hall mobility is not observed for these simpler structures, suggesting that interband scattering is responsible for the Anderson formula, $\cot\theta_H = \alpha T^2 + \beta$, that describes the Hall angle in the more complex high-T_c superconductors. Surprisingly, the appearance of a T^{-3} Hall mobility suggests that a previously undiscovered scattering mechanism influences the electrical transport properties of the high-T_c superconductors, but may be obscured by the multiband electron-electron scattering processes. As verification for this anomalous scattering, under plausible single-band transport conditions, we also have observed a conversion from the T^{-2} Hall mobility to the T^{-3} mobility just before the metal-insulator transition is reached in the superconducting (quenched) $YBa_2Cu_3O_{\sim 6.4}$ system ($T_c \sim 20$ K). This oxygen stoichiometry is expected to completely destroy the chain-derived bands, leaving only the hybridized plane-derived $Cu\,3d-O\,2p$ band intact [22, 23]. Finally, it would be interesting to determine the upper limit of temperature to which this unusual T^{-3} Hall mobility persists. The answer may help to determine whether or not phonon scattering plays any role in this anomalous temperature dependence.

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