Charge Transport Properties of Lightly-Doped Cuprates: Behavior of the Hall Coefficient

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Behavior of $\rho_{ab}(T)$ and $R_H(T)$ is presented for LSCO and YBCO single crystals in the lightly hole-doped antiferromagnetic region, with an emphasis on the $R_H(T)$ data. In both systems, $R_H$ is virtually constant at moderate temperatures and tends to increase at low temperatures. Since essentially the same behavior of $\rho_{ab}(T)$ and $R_H(T)$ is observed in both LSCO and YBCO, we discuss that the in-plane charge transport properties are universal among the cuprates in the lightly-doped regime and that the $R_H(T)$ data we obtained represent the genuine behavior of the Hall effect in this regime.

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1. INTRODUCTION

To elucidate the mechanism of high-$T_c$ superconductivity, it is indispensable to understand how the metallic charge conduction emerges and evolves from the parent Mott insulator as holes are doped to the two-dimensional CuO$_2$ planes. Fueled by this motivation, we have been intensively studying the lightly-doped region of the cuprates in the past few years, using high-quality single crystals of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) and YBa$_2$Cu$_3$O$_y$ (YBCO). During the course of our research, it has become evident that in the lightly-doped region, which is normally called “antiferromagnetic insulating regime”, doped holes are surprisingly mobile and show metallic transport at moderate temperatures in sufficiently clean single crystals, even when the in-plane resistivity significantly exceeds the Mott-Ioffe-Regel limit for metallic 2D transport. Moreover, the in-plane motion of the holes is found to be almost completely insensitive to the establishment of the long-range antiferromagnetic (AF) order, although at low temperatures holes show strong localization that can be characterized by a variable-range-hopping behavior.
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Such anomalous metallic transport at moderate temperatures suggests that charges are *mesoscopically segregated* from magnetic domains to form a self-organized network of hole-rich paths, which can be viewed as a nematic or isotropic phase of fluctuating charge stripes.

It has been reported that the Hall resistivity of La$_{1.4-x}$Nd$_{0.6}$Sr$_x$CuO$_4$ (LNSCO) tends to disappear upon transition into the static stripe phase, and Noda et al. has claimed that this is due to the intrinsically 1D nature of the charge transport in the stripe phase. Thus, given that the charge transport in the lightly-doped cuprates appears to be largely governed by the stripes, it is natural to ask how the Hall coefficient $R_H$ behaves in the lightly-doped region. Also, it was recently reported by Wang and Ong that in a specially-treated samples of YBCO in the AF regime, the Hall coefficient tends to vanish below the Néel temperature $T_N$. Such observation was proposed to be due to an intrinsic particle-hole symmetry in the system, which may well be related to the charge stripes or some other peculiarities.

Therefore, it is important to scrutinize the intrinsic behavior of $R_H$ in the lightly-doped AF regime of the cuprates. In this paper, we show our data of $R_H(T)$ for lightly-doped LSCO and YBCO, and discuss how we can sort out the genuine behavior of the Hall coefficient in the lightly-doped cuprates.

2. EXPERIMENTAL

The high-quality LSCO single crystals are grown by the traveling-solvent floating-zone (TSFZ) technique. The LSCO crystals are carefully annealed to remove excess oxygen, which is particularly important for lightly-doped samples in ensuring that the hole doping is exactly equal to $x$. The clean YBCO crystals are grown in Y$_2$O$_3$ crucibles by a conventional flux method to exclude the conductivity contribution from the Cu-O chains, which run along the $b$-axis in YBCO with $y \geq 6.35$, the crystals are detwinned at temperatures below 270°C with an uniaxial pressure of ~0.1 GPa, and the resistivity is measured along the $a$-axis in YBCO. The detwinning is done after the oxygen contents are tuned to the desired values by proper annealing. Note that the detwinning is not necessary for YBCO crystals with $y \leq 6.30$, where the system has tetragonal crystal symmetry. The in-plane resistivity $\rho_{ab}$ and the Hall coefficient $R_H$ are measured using a standard ac six-probe method. The Hall effect measurements are done by sweeping the magnetic field to $\pm 14$ T at fixed temperatures stabilized within ~1 mK accuracy. The Hall coefficients are always determined by fitting the $H$-linear Hall voltage in the range of $\pm 14$ T, which is obtained after subtracting the magnetic-field-symmetrical magnetoresistance component caused by small misalignment of
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the voltage contacts. Since we always check for the linearity of the Hall voltage for each magnetic-field sweep, we are confident that the Hall data shown here are free from any adverse effect of superconducting fluctuations, which could affect the results when samples contain a superconducting minor phase.

3. REMARKS ON YBCO SAMPLE PREPARATION

It should be emphasized that the oxygen arrangement in the Cu-O chain layers of YBCO is largely variable, which causes complications to the study of YBCO. For example, for a given $y$ the actual hole doping can differ depending on the arrangement of the O atoms, and the O atoms in the Cu-O chains can rather easily rearrange at room temperature; this causes the well-documented “room-temperature (RT) annealing effect"\(^3\), with which the $T_c$ of heavily underdoped, but still superconducting, sample slowly increases in several-day time scale. Long-time annealing at $80 - 150^\circ$C can enhance short-range oxygen ordering, but can never remove the intrinsic randomness unless $\delta$ ($= 7 - y$) is exactly 1/2 (for which well-ordered ortho-II phase can be obtained). For this work (and in all of our previous works\(^1,2,5,15,16,17\)), the crystals are always quenched at the end of the high-temperature annealing (which tunes the oxygen content) and detwinning is performed at temperatures below $220^\circ$C after the annealing. The samples are left at room temperature for at least a week for the local oxygen arrangement to equilibrate; therefore, the oxygen atoms on the chain sites are locally ordered (because of the RT annealing) but macroscopically uniform (because of the quenching)\(^2\). This procedure ensures very good reproducibility of the transport properties, as has been demonstrated in Ref.\(^1\) and gives a sharp superconducting transition\(^1\) (for $y > 6.35$) or a sharp Néel transition\(^2\) (for $y < 6.35$), both of which are hallmarks of uniform hole distribution in the sample. Also, we have never observed that a change in the annealing condition (including the RT annealing) gives rise to a change in residual resistivity; this implies that the randomness in the chain layers does not work as a strong disorder for the in-plane charge transport, which is understandable because the Cu-O chain layers are separated from the CuO$_2$ planes by the apical-oxygen layers and thus are rather far ($\sim4$ Å) away from the planes.

4. RESULTS

Figure 1 shows the in-plane resistivity $\rho_{ab}$ and the Hall coefficient $R_H$ of lightly-doped LSCO single crystals ($x = 0.01 - 0.03$). Note that the $x$
Fig. 1. Temperature dependences of (a) $\rho_{ab}$ and (b) $R_H$ of lightly-doped LSCO single crystals ($x = 0.01 - 0.03$). In panel (b), the values of $R_H^\text{min}$ expected for ordinary single-band metal with the three hole concentrations (0.01, 0.02, and 0.03 per Cu) are shown by solid lines.

$= 0.01$ sample shows long-range Néel order below 240 K (Ref. 3). One can easily see in Fig. 1(a) that the $\rho_{ab}(T)$ data of all the samples show metallic behavior ($d\rho_{ab}/dT > 0$) at moderate temperatures; in particular, in the $x = 0.01$ sample $\rho_{ab}(T)$ is completely insensitive to the Néel ordering and keeps its metallic behavior well below $T_N$. This insensitivity of the in-plane charge transport to Néel ordering may not be so surprising, because the large $J$ ($\sim 0.1$ eV) causes the antiferromagnetic correlations to be well established in the CuO$_2$ planes from above $T_N$ (Ref. 18).

Figure 1(b) shows the corresponding $R_H(T)$ data for the three concentrations. Note that $R_H$ is essentially temperature independent in the temperature range where the metallic behavior of $\rho_{ab}(T)$ is observed, which is exactly the behavior that ordinary metals show. Moreover, $R_H$ agrees well with the value $R_H^\text{min}$ expected for ordinary single-band metal with the nominal hole concentration of $x$ per Cu (i.e., $R_H^\text{min} \equiv V_0/\text{ex}$ where $V_0$ is the unit volume per Cu). At low temperature where disorder causes the holes to localize, $R_H(T)$ reflects the localization (i.e., decrease in the number of mobile holes) and tends to diverge, which is also an ordinary behavior for disordered metals with low carrier density. Therefore, in the lightly-doped LSCO, the behavior of $R_H$ appears to be a conventional one for disordered metals; this is quite opposite to the behavior in the superconducting regime,
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Fig. 2. Temperature dependences of $\rho_{ab}$ (left axis) and $R_H$ (right axis) of slightly-doped YBCO at $y = 6.22$. The value of $R_H^0$ expected for ordinary single-band metal with hole concentration of 0.005 per Cu is shown by solid line.

where $R_H$ significantly departs from $R_H^0$ and shows a strong temperature dependence.

Figure 2 shows $\rho_{ab}(T)$ and $R_H(T)$ data of a very lightly hole-doped YBCO single crystal ($y = 6.22$); remember that in YBCO the hole doping onto the CuO$_2$ planes starts above $y \simeq 6.20$. The behavior of both quantities is essentially the same as that in lightly-doped LSCO: $\rho_{ab}(T)$ shows a metallic behavior at moderate temperatures and is insensitive to the Néel ordering (which occurs above 300 K for this composition); $R_H$ is only weakly temperature dependent in the temperature range where the metallic $\rho_{ab}(T)$ is observed, and its magnitude is consistent with the expected $R_H^0$ value (we estimate the hole concentration per Cu, $p$, is less than 0.01 and probably around 0.005 for this composition). Also, $R_H(T)$ reflects the charge localization and tends to increase at low temperature. Therefore, our data indicate that the in-plane charge transport in the lightly-doped region is essentially universal among LSCO and YBCO, and that there is no hint of vanishing $R_H$ in the AF state.

For YBCO, we also show $\rho_a(T)$ and $R_H(T)$ data for higher, but still heavily underdoped, concentrations (Fig. 3); note that the samples for $y = 6.35$, 6.45, and 6.50 are detwinned single crystals. We estimate the $p$ value for $y = 6.35$ to be around 0.03, and in Fig. 3(b) one can see that the measured $R_H$ value at moderate temperatures are somewhat lower than, but
Fig. 3. Temperature dependences of (a) $\rho_\alpha$ and (b) $R_H$ of lightly-doped YBCO single crystals ($y$ = 6.22, 6.35, 6.45, and 6.50). In panel (b), the values of $R_H^n$ expected for ordinary single-band metal with hole concentrations of 0.005 and 0.03 per Cu are shown by solid lines.

close to the expected $R_H^n$ value for its hole concentration, just as the case of LSCO at $x = 0.03$. (Remember that in YBCO the actual $p$ value for a given $y$ varies depending on the oxygen arrangement as already mentioned, and thus the estimation of $p$ in YBCO can never be exact.)

5. DISCUSSIONS

5.1. Hall Coefficient and Stripes

As is mentioned in the Introduction, it has been shown that the Hall effect tends to disappear in LNSCO in the static stripe phase. Although such a result appears to naturally indicate the 1D behavior at first sight, one should note that the quasi-1D motion itself does not necessarily drive the Hall coefficient to zero. The quasi-1D confinement indeed suppresses the transverse (Hall) current induced by the magnetic field, but the same large transverse resistivity restores the finite Hall voltage, because of the relation $R_H \sim \sigma_{xy}/\sigma_{yy}\sigma_{xx}$. For the same reason, for example, the strong charge confinement in the CuO$_2$ planes in the cuprates does not prevent generation of the Hall voltage along the $c$-axis when $H$ is applied along the $ab$ plane. The anomaly in the Hall effect in LNSCO is likely to be caused
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by the peculiar arrangement of stripes which alter their direction from one CuO$_2$ plane to another, thereby avoiding $\sigma_{yy}$ to vanish. On the other hand, the unidirectional stripes that exist in pure LSCO in the lightly-doped region may keep the Hall coefficient unchanged. Also possible is the finite Hall resistivity being caused by the transverse (sliding) motion of the stripe as a whole; in fact, very recent optical conductivity measurements of lightly-doped LSCO showed that the sliding degrees of freedom are important for the realization of the metallic transport in this system. Therefore, even when the in-plane charge transport is essentially governed by the stripes in the lightly-doped cuprates, it is not at all surprising that $R_H$ does not vanish and shows rather conventional behavior.

5.2. Antiferromagnetic YBCO

As is shown in Fig. 3(b), $R_H(T)$ of our quenched YBCO samples does not exhibit any pronounced peak, but rather shows an upturn with decreasing temperature when the resistivity displays an insulating behavior; this result is in clear disagreement with the result reported in Ref. [1], which was obtained for samples annealed at 150$^\circ$C for 3 weeks. In 1992, Jorgensen’s group at Argonne National Laboratory intensively studied the effect of long-time annealing of YBCO; since it had already been known that in quenched samples the tetragonal-to-orthorhombic transition occurs at $y \simeq 6.35$, they carefully studied the samples which showed orthorhombicity down to $y = 6.25$ after a long-time annealing. They concluded that such orthorhombicity was a result of a macroscopic phase separation into tetragonal and orthorhombic phases; furthermore, they explicitly showed that this macroscopic phase separation took place when samples with $y$ of around 6.3 were annealed at low temperature for a long time. The mechanism of this phase separation can be understood by looking at the phase diagram (such as that in Ref. [23]); for example, when a sample is at $y = 6.30$ and is annealed at 150$^\circ$C for a long time, the proximity of the orthorhombic phase (which is stabilized above $y \simeq 6.4$ at this temperature and apparently has a lower free energy) in the phase diagram causes oxygen-rich domains with the orthorhombic symmetry to be formed and the rest of the sample is left to be oxygen-poor and tetragonal, leading to a macroscopic phase separation.

On the other hand, as the neutron scattering works have shown [22, 23], the quenched samples have no such problem as the macroscopic phase separation. In fact, the $R_H(T)$ data in Fig. 2 are essentially consistent with the $R_H(T)$ behavior in lightly-doped LSCO, which gives confidence that the $R_H(T)$ data we obtained in the antiferromagnetic YBCO represent the genuine behavior.
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of the Hall coefficient of the lightly-doped cuprates. Since other in-plane transport properties show striking similarities between LSCO and YBCO in the lightly-doped region, it is actually more natural to see the Hall effect to behave similarly in the two systems, than to see quite different behaviors.

6. SUMMARY

High-quality data of $\rho_{ab}(T)$ and $R_H(T)$ are shown for LSCO and YBCO single crystals in the lightly-doped region. The presentation is focused on $R_H(T)$, which exhibits almost temperature-independent value in the temperature range where $\rho_{ab}(T)$ shows a metallic behavior. The value of $R_H$ in this nearly-temperature-independent regime agrees rather well with the value expected from the hole concentration for ordinary single-band metal. Notably, such behavior of $R_H(T)$ is observed in both LSCO and YBCO in the lightly-doped region, suggesting that the in-plane charge transport properties are universal among cuprates in the AF regime; this gives us confidence that the $R_H(T)$ data we obtained represent the genuine behavior of the Hall effect in the lightly-doped cuprates.

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