Mobility of the Doped Holes and the Antiferromagnetic Correlations in Underdoped High-T_c Cuprates

Yoichi Ando, A. N. Lavrov, Seiki Komiya, Kouji Segawa, and X. F. Sun

Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan

The emergence and the evolution of the metallic charge transport in the $La_{2-x}Sr_xCuO_4$ system from lightly to optimally doped samples ($x = 0.01-0.17$) are studied. We demonstrate that in high-quality single crystals the in-plane resistivity shows a metallic behavior for *all* values of x at moderate temperatures and that the hole mobility at 300 K changes only by a factor of 3 from $x = 0.01$ to 0.17, where its x dependence is found to be intriguingly similar to that of the inverse antiferromagnetic correlation length. We discuss an incoherent-metal picture and a charged-stripe scenario as candidates to account for these peculiar features.

The underdoped region of the high-T_c cuprates has attracted significant attention in recent years. Quite peculiar features such as pseudogap [1], $\log(1/T)$ insulating behavior [2], and charged-stripe instability [3,4] have been discovered in the underdoped region, and those features are expected to bear essential clues to elucidating the origin of the high-T_c superconductivity. The $La_{2-x}Sr_xCuO_4$ (LSCO) system has been most frequently used for systematic studies of the moderately to heavily underdoped region of the cuprates, because of the accessibility to the low-doped region. Neutron scattering and photoemission experiments on the LSCO system have provided detailed knowledge about the evolution of the spin [4,5] and electronic [6,7] structures as holes are doped to the parent Mott insulator; however, how the metallic charge conduction emerges in this doped Mott insulator is still poorly understood. It has been proposed theoretically that, because of the strong magnetic correlations in cuprates, the metallic conduction may be realized through spin-charge separated quasiparticles [8,9], charged stripes [10,11], incoherent quasiparticles [12], etc. To fully understand the transformation from the Mott insulator to a metal in cuprates, systematic studies of the transport properties in the heavily underdoped region are highly desirable.

In the early days of high-T_c research, the transport properties of the heavily underdoped LSCO were studied using polycrystals, thin films, and flux-grown crystals [5]. These works showed that the charge transport became insulating in the nonsuperconducting samples with the variable-range-hopping (VRH) conductivity at low temperatures, while at high temperatures the transport could be metallic already in the spin-glass regime [5]. However, there has been no systematic measurement of the transport properties of the high-quality single crystals in the heavily underdoped region, which have become available with the development of the traveling-solvent floating-zone (TSFZ) technique [13]. Since one can achieve high uniformity of the Sr concentration and high purity in the TSFZ-grown crystals, it has become possible to study, for example, the

doping dependence of the mobility of the holes in clean and well-controlled crystals down to the heavily underdoped antiferromagnetic region without being bothered by extraneous disorder.

In this Letter, we report the in-plane resistivity ρ_{ab} and the Hall coefficient R_H measurements of a series of TSFZ-grown high-quality LSCO crystals that cover the whole underdoped region, from $x = 0.01$ to 0.17. It is found that in high-quality crystals the behavior of $\rho_{ab}(T)$ is metallic ($d\rho_{ab}/dT > 0$) at moderate temperatures for all values of x, even in the Néel state at $x = 0.01$; since the magnitude of $\rho_{ab}(T)$ in the lightly doped region is far above the Mott limit for metallic conduction in two-dimensional (2D) metal, this is a strong manifestation of the "bad metal" [14] behavior. Moreover, we observe that the mobility of doped holes in this "metallic" regime shows a doping dependence similar to that of the inverse antiferromagnetic (AF) correlation length, ξ_{AF}^{-1}. Notably, the absolute value of the mobility changes only by a factor of 3 from $x = 0.01$ to 0.17 at 300 K. These results demonstrate the peculiar nature of the charge transport in the cuprate that is a doped Mott insulator with strong magnetic correlations and give essential clues to elucidate the origin of the bad metal behavior in cuprates. To corroborate the result on LSCO, we also show the $\rho_a(T)$ data of $YBa_2Cu_3O_y$ (YBCO) for a similarly wide range of doping.

The clean single crystals of LSCO are grown by the TSFZ technique and are carefully annealed to remove excess oxygen, which ensures that the hole doping is exactly equal to x. The crystallographic axes are determined by the Laue analysis and then the samples are shaped into thin platelets with the ab planes parallel to the wide face, where an error in the axes' directions is less than 1°. The YBCO crystals are grown in Y_2O_3 crucibles by a conventional flux method and are detwinned at temperatures below 220 °C with a uniaxial pressure of ~0.1 GPa after the oxygen content is tuned to the desired doping with careful annealing and quenching [15]. The resistivity in YBCO is measured along the a axis to exclude the conductivity

contribution from CuO chains, which run along the b axis. Note that the detwinning is not necessary for YBCO crystals with $y < 6.40$, which have tetragonal symmetry.

The in-plane resistivity ρ_{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 ± 14 T at fixed temperatures stabilized within ~ 1 mK accuracy [16]. The uncertainty in the absolute magnitude of ρ_{ab} and R_H is minimized [15] by using relatively long samples (voltage contact separation is typically ~ 1 mm for YBCO and ~ 1.5 mm for LSCO), painting narrow contact pads with the width of $50-80$ μm, and accurately determining the crystal thickness by measuring the weight with 0.1-μg resolution; total errors in absolute values of ρ_{ab} and R_H are less than 10% and 5%, respectively. In our crystals, ρ_{ab} is very reproducible (we always measure several crystals for each composition), and its absolute value is among the smallest ever reported for each composition.

Figure 1 shows the temperature dependences of ρ_{ab} for the LSCO crystals, with the vertical axis in the logarithmic scale. One may immediately notice that ρ_{ab} in the moderate-temperature range show metallic behavior ($d\rho_{ab}/dT > 0$) for *all* values of x. It is particularly intriguing to note that in the $x = 0.01$ sample $\rho_{ab}(T)$ keeps its metallic behavior well below the Néel temperature T_N (which is 240 K; see Fig. 2 inset). This clearly demonstrates that the in-plane charge transport is insensitive to the long-range magnetic order, which may not be surprising because the large J (~ 0.1 eV) causes the antiferro-

magnetic correlations to be well established in the CuO_2 planes far above T_N [5].

To examine the detailed doping dependence of the charge transport, it is useful to look at the conductivity per charge, namely, the mobility of the doped holes. In Fig. 2, we plot the temperature dependences of $n_h e \rho_{ab}$ for various x, where e is the electronic charge and n_h is the nominal hole concentration given by $2x/V$ [unit cell V ($\simeq 3.8 \times 3.8 \times 13.2$ Å^3) contains two CuO_2 planes in LSCO]. This product, $n_h e \rho_{ab}$, corresponds to the inverse mobility μ^{-1} of the doped holes. Note that it is probably better to use $n_h e$ ($= 2ex/V$) than to use R_H^{-1} for the calculation of the conductivity per hole, because R_H in cuprates shows a strong temperature dependence that is not caused by a change in the density of mobile holes. One can see in Fig. 2 that the slope of $n_h e \rho_{ab}(T)$ at 300 K depends only weakly on x and the absolute value of μ^{-1} changes only by a factor of 3 at 300 K; the similarity of the moderate-temperature $\mu^{-1}(T)$ curves and the rather small change in their magnitude hint at the possibility that the metallic charge transport is governed by essentially the same mechanism from $x = 0.01$ to 0.17.

Figure 3 shows the x dependence of μ^{-1} at 300 K. The change in μ^{-1} is smooth and is relatively small, which is rather surprising in view of the superconductor-insulator transition occurring at $x \approx 0.05$. In Fig. 3, we also plot the x dependence of the AF correlation length ξ_{AF}, which is known to show a similarly smooth change with x; $\xi_{AF}(x)$
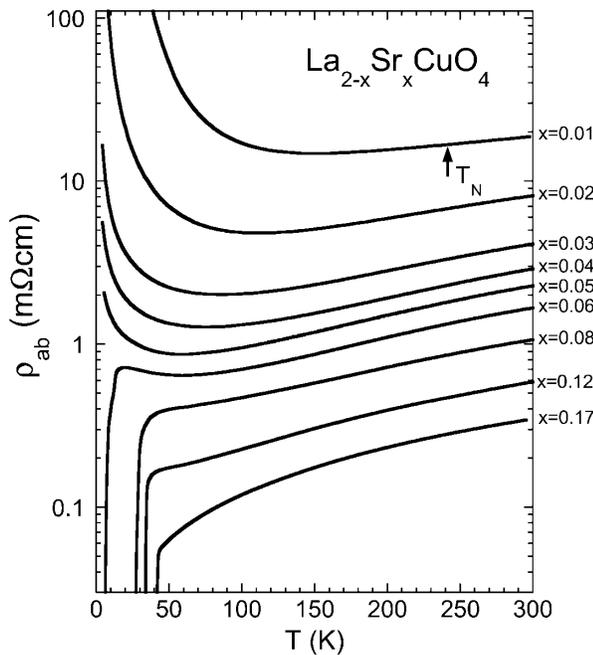

FIG. 1. Temperature dependences of ρ_{ab} in TSFZ-grown La$_{2-x}$Sr$_x$CuO$_4$ single crystals.
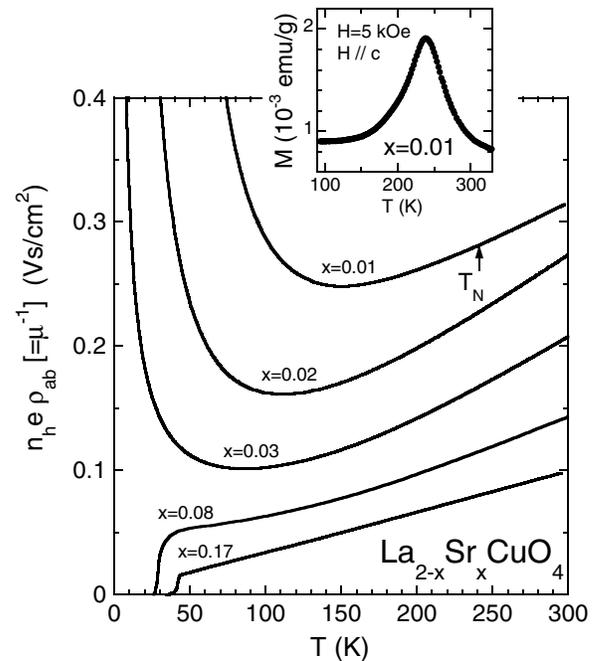

FIG. 2. Temperature dependences of the inverse mobility, $n_h e \rho_{ab}$, of the LSCO crystals. Inset: magnetization of a large La$_{1.99}$Sr$_{0.01}$CuO$_4$ single crystal (93 mg) from which the samples for ρ_{ab} measurements were cut; the peak in $M(T)$ corresponds to the Néel transition.
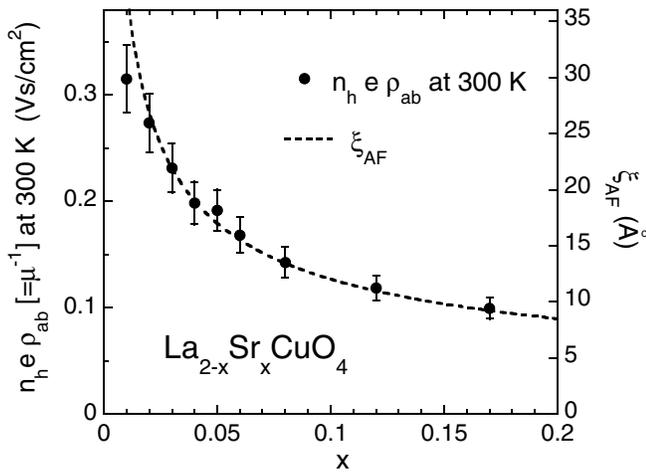
FIG. 3. x dependence of the inverse mobility (left axis) of the LSCO crystals. The dashed line shows the x dependence of ξ_{AF} (right axis), which is reported to be $3.8/\sqrt{x}$ Å by neutron experiments [5].
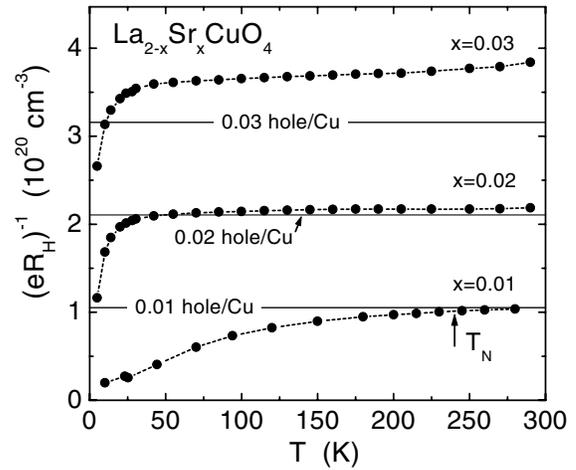

FIG. 4.   The apparent density of carriers $n = (eR_H)^{-1}$ for the three lightly doped LSCO crystals; solid lines indicate the nominal hole density n_h for each doping.

is reported to be described by $3.8/\sqrt{x}$ Å [5], the average separation between the doped holes. (Note that ξ_{AF} is almost temperature independent below ~ 300 K except for $x = 0.01$ [5].) It is striking that both μ^{-1} and ξ_{AF} show very similar x dependence; in particular, both show only a factor of 2 or 3 change from $x = 0.02$ to 0.17 despite the significant change in the ground state. The similarity of the x dependences of μ^{-1} and ξ_{AF} suggests that the charge transport in the underdoped cuprates is inherently related to the magnetic correlations in the background spins. It is worthwhile to mention that the magnitude of μ of LSCO is $3-10$ cm^2/V s at 300 K in the whole doping range studied, and these numbers are close to those of typical metals [e.g., $(ne\rho)^{-1}$ of iron is 4 cm^2/V s at 273 K [17]].

We note that the metallic behavior of $\rho_{ab}(T)$ at moderate temperatures observed in the heavily underdoped samples ($x \leq 0.05$) indicates that the system is not an insulator (or a semiconductor) with a well-defined gap. The low temperature insulating behavior in these samples is consistent with the VRH behavior as reported before [5], which also suggests that there is a developing band of electronic states at the Fermi energy E_F. Therefore, given that the chemical potential is pinned in the middle of the Mott-Hubbard gap [7], the resistivity data tell us that with only 1% of doping a band is created near E_F within the gap of the parent cuprate, and the system starts to show bandlike transport (with disorder). The Hall coefficient data also support this picture; namely, as in ordinary metals, the apparent hole density $n \equiv (eR_H)^{-1}$ of the heavily underdoped samples is essentially temperature independent in the temperature range where the metal-like behavior of $\rho_{ab}(T)$ is observed, as shown in Fig. 4 for $x = 0.01-0.03$. Note that, at low doping, n agrees well with the nominal hole density n_h at moderate temperatures, which means that all the doped holes are moving and contributing to the Hall effect.

The above results on LSCO are essentially reproduced in YBCO. Figure 5 shows the temperature dependences of $n_h e \rho_a$ for clean untwinned YBCO crystals with various oxygen contents. Although the determination of hole concentrations in YBCO is not as straightforward as in LSCO, one may estimate [18] the hole number per Cu to be around 0.02, 0.05, and 0.17 for samples with $y = 6.30$, 6.45, and 6.95, respectively. These doping levels cover the range from the AF region ($T_N = 230$ K for $y = 6.30$ [16]) to an optimally doped superconductor with $T_c = 93.4$ K. As is the case with LSCO, the metallic behavior is insensitive to the establishment of the AF state at T_N, and μ^{-1} at 300 K changes only by a factor of 2 from the AF "insulator" to the optimally doped superconductor. Note that both LSCO and YBCO show similar μ of around 10 cm^2/V s at moderate temperatures, which suggests that the hole

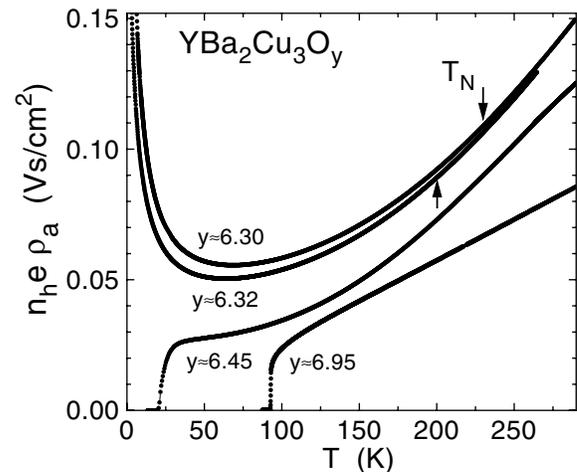FIG. 5.   Temperature dependences of the inverse mobility, $n_h e \rho_a$, of untwinned YBa$_2$Cu$_3$O$_y$ crystals. The Néel temperatures [16] for lightly doped ($y = 6.30$ and 6.32) crystals are indicated by arrows.

mobility is almost universal among the cuprates. Also, though not shown here, the $R_H(T)$ data of the lightly doped YBCO are essentially similar to those of LSCO and are consistent with the data published recently [19].

In the above results, one of the notable findings is that a metallic behavior of $\rho_{ab}(T)$ at moderate temperatures is already established even for 1% of hole doping. Although such a metallic behavior suggests a Boltzmann band transport for the doped holes, the *magnitude* of ρ_{ab} is so large that $k_F l$ would be only 0.1, which strongly violates the Mott limit for the metallic transport (k_F is the Fermi wave number and l is the mean free path) [20], and therefore the charge transport cannot be caused by the motion of ordinary quasiparticles in a 2D electronic system. In this sense, this is a strong manifestation of the bad metal [14] behavior at low doping, although the bad metals have often been discussed in conjunction with the absence of resistivity saturation at high temperature [14]. Our results on the behavior of μ^{-1} (Figs. 2 and 3) suggest that the mechanism that causes the bad metal behavior in cuprates is likely to be fundamentally related to the background AF correlations and that the same mechanism may well govern the charge transport up to $x = 0.17$. Below we discuss two possibilities for the charge transport mechanism that may be consistent with the peculiarities observed here.

One possible picture is the "incoherent metal" [6,12], where E_F is smaller than $k_B T$ and thus the charge transport is essentially a diffusion process of classical particles; since the charge dynamics is diffusive and thus is not strictly a band transport, the Mott limit is completely irrelevant in this case. Still, the peculiar T-linear resistivity may be expected in an incoherent metal [12], because the large magnetic energy scale causes the system to be quantum critical. Recent photoemission results on LSCO also suggest that the incoherent metallic state may emerge in the lightly doped region from the parent Mott insulator [6]. However, it may be difficult to understand the apparent connection between μ^{-1} and ξ_{AF} in the incoherent-metal picture.

Another possibility is that the phase segregation on the mesoscopic scale changes the effective volume relevant to the charge transport. In this case, both the apparent violation of the Mott limit and the insensitivity of the mobility to the change in x can be naturally understood. For example, when the charged stripes are formed, metallic transport through the quasi-one-dimensional charge system may be possible [10,11]. Since the charged stripes are rather decoupled from the magnetically ordered regions that separate them, the metallic behavior may only weakly depend on x; however, the apparent correlation between μ^{-1} and ξ_{AF} indicates that the charge transport is influenced by the rigidity of the magnetic correlation in the magnetic domains, which probably means that the transverse fluctuation of the stripes in the AF environment must be significant within this picture. In fact, a recent theory predicts [21] that the transverse fluctuation of the charged stripes gives rise to "electronic liquid crystals," which can behave like a 2D metal.

In summary, we show that the inverse mobility μ^{-1} at moderate temperatures shows a metallic temperature dependence in the whole underdoped region down to $x = 0.01$ and that the magnitude of μ^{-1} at 300 K changes only by a factor of 3 from $x = 0.01$ to 0.17 in high-quality single crystals. Moreover, it is found that the x dependence of μ^{-1} at 300 K appears to be intriguingly similar to that of ξ_{AF} in the whole underdoped region. These features suggest that the *same* mechanism governs the charge transport from $x = 0.01$ to 0.17 and that such a mechanism is fundamentally related to the background AF correlations. It is discussed that both an incoherent metal or charged stripes can basically explain the unusual metallic charge transport, though the apparent correlation between μ^{-1} and ξ_{AF} can be more easily understood within the stripe scenario.

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