Evolution of the Hall Coefficient and the Peculiar Electronic Structure of the Cuprate Superconductors

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Although the Hall coefficient R_H is an informative transport property of metals and semiconductors, its meaning in the cuprate superconductors has been ambiguous because of its unusual characteristics. Here we show that a systematic study of R_H in $La_{2-x}Sr_xCuO_4$ single crystals over a wide doping range establishes a qualitative understanding of its peculiar evolution, which turns out to reflect a two-component nature of the electronic structure caused by an unusual development of the Fermi surface recently uncovered by photoemission experiments.

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During the past 17 years after the high-T_c superconductivity was discovered in cuprates, virtually all measurable properties of their "normal state," the state in the absence of superconductivity, have been studied to understand the *stage* for novel superconductivity. However, there is yet no established picture for even such basic properties as the resistivity and the Hall coefficient [1], not to mention other more elaborate properties. The Hall coefficient R_H of conventional metals is independent of temperature and signifies the Fermi surface (FS) topology and carrier density, but in cuprates R_H shows strong, sometimes peaked, temperature dependences as well as a complicated doping dependence. An advance in understanding came when Chien, Wang, and Ong found [2] that the cotangent of the Hall angle, $\cot\Theta_H$ (which is the ratio of the in-plane resistivity ρ_{ab} to the Hall resistivity ρ_H), approximately shows a simple linear-in-T^2 behavior, which suggests the existence of a quasiparticle-relaxation rate that changes as $\sim T^2$. However, while it appears that the Hall problem in cuprates can be simplified when analyzed in terms of $\cot\Theta_H$, it was argued by Ong and Anderson [3] that $\cot\Theta_H$ is after all a derived quantity, and the central anomaly resides in the directly measured quantities ρ_{ab} and R_H.

In this Letter, we address the notoriously difficult problem of the Hall effect with the recent knowledge on the physics of *lightly doped cuprates* and the peculiar evolution of the FS recently elucidated by the angle-resolved photoemission spectroscopy (ARPES) experiments [4,5]. We first show that the behavior of R_H and ρ_{ab} in the lightly doped cuprates mimics rather well the behavior of a conventional Fermi liquid, and discuss that this behavior signifies the physics on the "Fermi arc," a small portion of the FS near the Brillouin-zone diagonals. We then discuss that the peculiar hole-doping dependence and the temperature dependence of R_H reflect a gradual participation of the "flatband" near $(\pi, 0)$ of the Brillouin zone, which brings about a sort of two-band nature to the transport. The measurements of R_H and ρ_{ab} using a standard six-probe method are done on high-quality single crystals of $La_{2-x}Sr_xCuO_4$ (LSCO) and $YBa_2Cu_3O_y$ (YBCO), the details of which have been described elsewhere [6].

In slightly hole-doped LSCO and YBCO, which are usually considered to be antiferromagnetic insulators, it was demonstrated [6] that the charge transport shows a surprisingly metallic behavior with a hole mobility comparable to that of optimally doped superconductors at moderate temperatures. Detailed ARPES measurements were subsequently performed on lightly doped LSCO [5] and YBCO [7], which revealed that only patches of FS, called "Fermi arcs" [8], are observed at the zone diagonals, where quasiparticlelike peaks were detected in harmony with the transport results. The ARPES results indicate that, for some reason, a significant fraction of the large FS (that is observed in optimally doped cuprates [9]) is destroyed and the remaining small portion is responsible for the metallic transport. Thus, by looking at the transport properties of the lightly doped cuprates, one can gain insight into the physics of the Fermi arc, whose origin is currently under debate [10].

Figures 1(a) and 1(b) show an example of the behavior of $\rho_{ab}(T)$ and $R_H(T)$ in a lightly doped cuprate: Here the data are for YBCO with $y = 6.30$ (hole doping of about 3% per Cu [6]), which is an antiferromagnet with the Néel temperature of 230 K [11]. As we have reported previously [6,12], R_H is virtually T independent (as in conventional metals) at moderate temperatures in the lightly doped samples; as a result, ρ_{ab} and $\cot\Theta_H$ have the same T dependence [see Figs. 1(c) and 1(d)], which, intriguingly, is most consistent with $\sim T^2$ and not with $\sim T$. This implies that the relaxation rate of the "quasiparticles" on the Fermi arc changes as $\sim T^2$, which incidentally is the same as the behavior of conventional Fermi liquids. Note that the low-temperature upturn in ρ_{ab} and R_H is due to localization effects [13], which just obscure the intrinsic low-temperature behavior of the system.

We found that the T dependence of ρ_{ab} is consistent with $\sim T^2$ not only in lightly doped YBCO but also in lightly doped LSCO, as shown in Fig. 2(a) for $x = 0.02$;

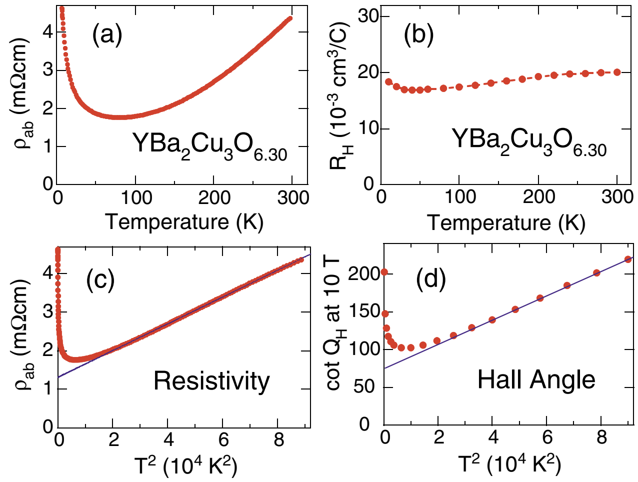

FIG. 1 (color online). Temperature dependences of transport properties of lightly hole-doped $YBa_2Cu_3O_{6.30}$. At this insulating composition, $\rho_{ab}(T)$ shows positive curvatures (a) and R_H is virtually T independent at moderate temperatures (b). It turns out that $\rho_{ab}(T)$ well obeys the T^2 law (c), which also holds for $\cot\Theta_H$ [$\sim \rho_{ab}/R_H$, (d)], suggesting that a Fermi-liquid-like T^2 scattering rate governs the transport on the Fermi arcs.

however, it should be noted that the temperature range for the T^2 law is a bit narrow [Fig. 2(a) shows the data up to 250 K] and $\rho_{ab}(T)$ deviates downwardly from the T^2 dependence at high temperatures. As is shown for $x = 0.08$ [Fig. 2(b)], the deviation tends to start from lower temperature at larger x. One possible way to interpret this deviation is to ascribe it to an increase in the density of states at the Fermi energy E_F with increasing T, which happens, for example, when a gap is filled in with T. In fact, the band structure of LSCO elucidated by ARPES [4,5] suggests such a possibility: In lightly doped LSCO a flatband [located near $(\pi, 0)$ of the Brillouin zone] lies below E_F and this band gradually moves up to E_F with increasing doping; therefore, if thermal activation causes some holes to reside on the flatband and to contribute to the conductivity, the high-temperature deviation from the T^2 behavior and its doping dependence can be understood, at least qualitatively.

The systematics of $R_H(T)$ and $\cot\Theta_H(T)$ of LSCO for a very wide range of doping ($x = 0.02$–0.25) is shown in Fig. 3. Let us first compare the robustness of the T^2 behavior in $\cot\Theta_H$ to that in ρ_{ab}: One can see in Fig. 3(b) that $\cot\Theta_H$ for both $x = 0.02$ and 0.08, where the behavior of ρ_{ab} was discussed, shows no high-temperature deviation from the T^2 behavior up to 300 K. This contrast between $\rho_{ab}(T)$ and $\cot\Theta_H(T)$ regarding the robustness of the T^2 law is qualitatively understandable if both ρ_{ab} and R_H reflect a change in the effective carrier density $n_{\rm eff}$ due to the temperature-dependent participation of the flatband, since such a change tends to be canceled in $\cot\Theta_H$; remember, $H\cot\Theta_H$ is equal to the inverse Hall mobility and, thus, would normally be free from a change in the carrier density. This observation suggests that the relative simplicity in the behavior of $\cot\Theta_H$ comes from its lack of a direct dependence on $n_{\rm eff}$, while both ρ_{ab} and R_H depend directly on $n_{\rm eff}$.

Thus, our data are most consistent with an emerging picture that a Fermi-liquid-like transport results from the quasiparticles on the Fermi arcs in lightly hole-doped cuprates, and the rest of the FS starts to contribute to the transport at higher doping and/or temperature. It should be noted, however, that there cannot be a real "Fermi liquid" on the Fermi arcs, because the large magnitude of ρ_{ab} in the lightly doped cuprates would indicate that the mean-free path of the electrons at E_F

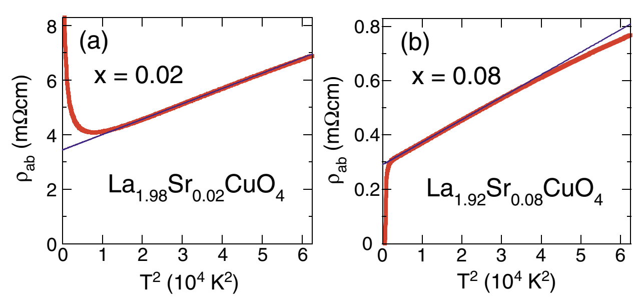

FIG. 2 (color online). Validity of the T^2 law in $\rho_{ab}(T)$ of LSCO single crystals. At a lightly doped composition $x = 0.02$ (a), the T^2 law (shown by a solid line) holds for the temperature range of 130–230 K, while at $x = 0.08$ (b) the range is 60–160 K.

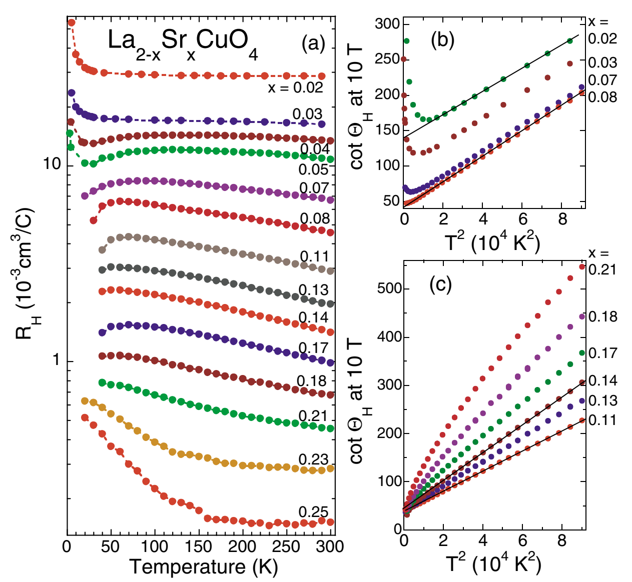

FIG. 3 (color online). Hall response in LSCO single crystals for a wide range of doping. (a) Variation of the T dependence of R_H for $x = 0.02$–0.25, all measured on high-quality single crystals; the x values are determined by the inductively coupled-plasma atomic-emission-spectroscopy analyses and are accurate within $\pm 5\%$. (b),(c) Plots of $\cot\Theta_H$ vs T^2 for representative x values; for selected data, solid lines emphasize the T^2 law in $\cot\Theta_H(T)$, which holds well for $0.02 \leq x \leq 0.14$.

is *shorter* than their de Broglie wavelength [6], which is impossible in a Fermi liquid; it was proposed that some self-organized inhomogeneity in the real space (such as charge stripes [14]) would offer a resolution to this apparent puzzle [6]. Also note that the patchy Fermi arcs which do not enclose a well-defined area cannot host a Fermi liquid. Therefore, the lightly doped cuprates are peculiar in that what appears to be conventional Fermi-liquid-like behavior characterizes the system which cannot really be a Fermi liquid.

In passing, we note that in Fig. 3(c) the T^2 law of $\cot\Theta_H$ holds very well up to optimum doping ($x \simeq 0.16$), but gradually breaks down when samples are overdoped; this observation confirms the trend previously noted by Hwang *et al.* [15] for polycrystalline samples. It is tempting to associate this change in the behavior of $\cot\Theta_H$ to the putative quantum phase transition [1] at optimum doping in LSCO, for which other transport properties also give indirect evidence [16,17]. Interestingly, similar change in the power-law behavior of $\cot\Theta_H$ has been observed to occur above $p \simeq 1/8$ (p is the hole doping per Cu) in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ [18], where the transition between "metal" and "insulator" under 60 T was found to lie also at $p \simeq 1/8$ [19]. It has been discussed that these crossover phenomena and some of the nonuniversalities may be attributed to competing orders [1,20]; in this regard, it is useful to mention that the asymmetries between hole-doped and electron-doped cuprates may also be due to some sort of competing orders [20].

Next we discuss the evolution of R_H with doping in more detail. It is instructive to plot eR_Hx/V vs T for various dopings (e is electron charge and V is unit volume per Cu); note that eR_Hx/V should be 1 if R_H simply signifies the nominal hole density, x/V. Figure 4(a) shows such a plot for LSCO at representative dopings. One can see that eR_Hx/V is actually 1 at around 100 K for $x =$ 0.02–0.07, but it tends to decrease from 1 with increasing temperature and this decrease starts from lower temperature as x becomes larger. This behavior suggests that on the Fermi arcs [Fig. 4(b)], which appear to govern the transport in these lightly doped samples at ~100 K, the carrier density is exactly equal to the nominal density of doped holes. As x is increased above 0.07, eR_Hx/V becomes smaller and never reaches 1; this trend is most likely related to the evolution of the flatband that we already mentioned, because this band touches E_F for $x \gtrsim$ 0.10 [4]. Therefore, it appears that the magnitude of R_H actually reflects the effective carrier density which becomes larger than the nominal hole density when the flatband starts to participate in the physics near E_F in addition to the Fermi arcs as doping and/or temperature is increased.

To further investigate the effect of temperature, we have measured R_H for $x = 0.02$ up to 900 K, and the resulting data [Fig. 4(c)] turn out to be surprisingly informative: they suggest that even at such low doping as $x = 0.02$ the flatband starts to participate in the Hall

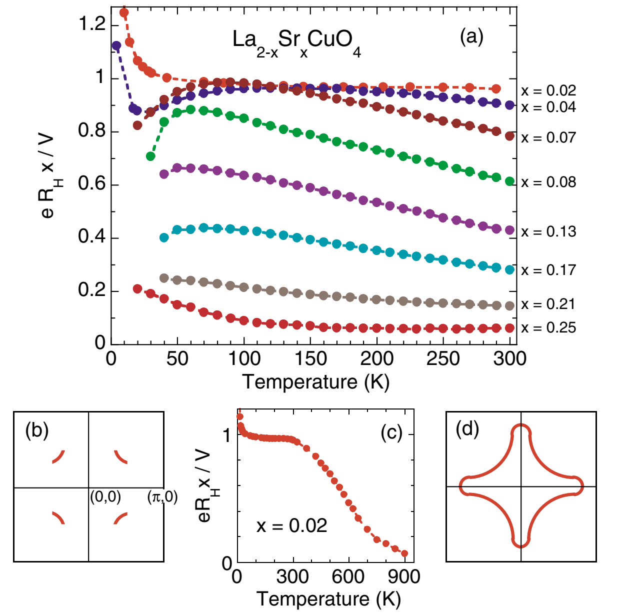

FIG. 4 (color online). Evolution of $R_H(T)$ and the Fermi surface in LSCO. (a) Temperature dependences of eR_Hx/V, which gives the ratio of measured Hall coefficient to that expected from nominal hole density, x/V, for representative x values; higher temperature data up to 900 K are shown for $x = 0.02$ (c). Schematic pictures of the Fermi arcs (b) and the electronlike closed FS (d) are also shown. The T-independent behavior of R_H observed at moderate temperatures for the two extremes, $x = 0.02$ and 0.25, measures the carrier density on the Fermi arcs and on the closed FS, respectively, and can be rather classically interpreted.

response above 300 K, which implies that the **k**-space patchiness melts away with thermal fluctuations and a full Fermi surface is eventually restored. Also informative is the behavior of R_H in our most overdoped sample, $x = 0.25$, where R_H is essentially T independent down to ~150 K. For overdoped LSCO, ARPES found [4] that the FS is "electronlike" with the shape depicted in Fig. 4(d); since this FS contains both positive- and negative-curvature parts, even within the conventional Boltzmann transport theory the value of R_H is determined by a rather complicated balance between the contributions from the two parts [21]. Remember that the positive (negative) curvature of the FS results in hole(electron)like Hall response. Therefore, one can interpret that the T-independent part of R_H for $x = 0.25$ is actually determined by the shape of the FS in a classical way, while the low-temperature upturn can be due to a development of the pseudogap [15] and/or some anisotropy in the relaxation rate on the FS [21]. This interpretation also naturally explains the sign change in the high-temperature value of R_H in further overdoped samples [15], for which ARPES showed that the positive-curvature part of the FS is gradually diminished [4], making electronlike Hall response to become dominant. Note that the precise T dependence of R_H for a given x is

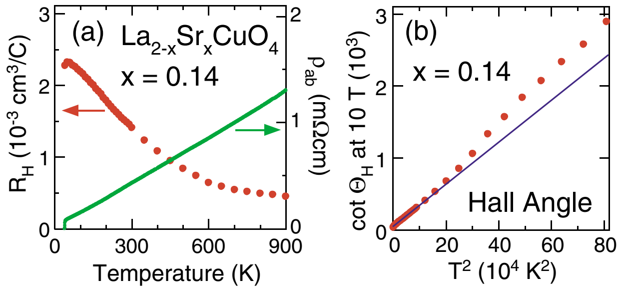

FIG. 5 (color online). (a) Temperature dependences of ρ_{ab} and R_H for LSCO at $x = 0.14$ up to 900 K. (b) T^2 plot of $\cot\Theta_H$, which demonstrates that the T^2 law breaks down above ~400 K.

likely to be governed both by the participation of the flatband and by the anisotropic relaxation rate in a complicated way, which is probably the reason why the Hall effect in the cuprates has been so difficult to be understood.

One may notice that a natural extension of the present argument would be that the T-linear resistivity usually observed near optimum doping may not necessarily be a sign of a T-linear relaxation rate, because $n_{\rm eff}$ may be changing with T. To address this issue, we show data for ρ_{ab}, R_H, and $\cot\Theta_H$ of LSCO at $x = 0.14$ up to 900 K in Fig. 5. One can see that R_H tends to saturate at high T (as it happens for $x = 0.25$) and the T^2 law in $\cot\Theta_H$ breaks down above ~400 K, both of which suggest that there is an electronic crossover with increasing temperature [this can be the pseudogap crossover (see Ref. [15])]. Therefore, while the T-linear resistivity is indeed observed up to 900 K [Fig. 5(a)], it is not very likely that $n_{\rm eff}$ stays constant and the relaxation rate is T linear up to 900 K. It is worthwhile to mention that the question of whether $n_{\rm eff}$ is changing with temperature or just the relaxation rate is changing is similar to the question regarding the interpretation of the optical conductivity $\sigma_1(\omega)$ of cuprates [22]; namely, the peculiar ω dependence of σ_1 can be interpreted either with a two-component model (simple Drude + midinfrared resonance) or with an extended Drude model (where the relaxation rate changes with ω). The former allows more electrons to respond to the electric field at higher energy, which bears similarities to our "two-band" picture (Fermi arc + flatband) for the dc transport.

Thus, although the detailed situation at optimum doping needs further clarification, the present results demonstrate that the physics of the normal state is phenomenologically simple in the two extremes, the lightly doped regime and the heavily overdoped regime; in particular, R_H has a classical meaning in these two extremes at moderate temperatures. The evolution from one to the other is a transition in the **k** space from the Fermi arc to a closed Fermi surface (patchiness to continuity), upon which the flatband appears to provide additional carriers and play a crucial role. Interestingly, increasing temperature also causes the same transition from patchiness to continuity. If the peculiar transport properties in the lightly doped cuprates are inherently related to a self-organized inhomogeneity in the real space [23] as some resent observations seem to indicate [6,11,13,24], the patchiness in the **k** space is likely to be the other side of the same coin. Therefore, a key to understanding the cuprates probably lies in the dual transition from patchiness to continuity in the **k** space and from inhomogeneity to homogeneity in the real space.

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