Evolution of charge carriers for transport in electron-doped cuprate superconductor $La_{1.89}Ce_{0.11}CuO_4$ thin films

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To reveal the evolution of charge carriers to the transport in $La_{1.89}Ce_{0.11}CuO_4$, we carefully measured the temperature and magnetic-field dependences of the Hall resistivity (ρ_{xy}) and longitudinal resistivity (ρ_{xx}). Two sign reversals of the Hall resistivity are observed in the normal state. In addition, the ρ_{xy} and ρ_{xx} show B^2 dependence in the temperature region of 40–50 K, similar to the behavior of the compensated metal; that is, the effective concentrations of electrons (n_e^*) and holes (n_h^*) are nearly equal. The possible origin of the sign reversal is attributed to the competition between n_e^*/n_h^* and τ_e/τ_h (the relaxation time ratio of electrons to holes), and the change of cyclotron orbits induced by the magnetic field is also considered.

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Electron-doped high-T_c cuprate superconductors have their own features in comparison with hole-doped ones, such as the T^2 dependence of the resistivity in the optimally and overdoped regions, the anomalous behavior of the Hall effect and magnetoresistivity, the two kinds of charge carriers, and so on.[1–3] So transport measurements should be an effective way to address the important issues of the cuprate superconductors, such as the development of the Fermi surface,[4,5] the existence of the pseudogap[6] and the quantum critical point,[7] etc. If such study can be done extensively, great progress will be achieved for the mechanism of high-T_c superconductivity. During the past years, the concept of two kinds of charge carriers and related transport properties became an attractive topic for electron-doped superconductors. The angle-resolved photoelectron spectoscopy experiment showed that in the electron-doped superconductor $(Nd,Ce)_2CuO_4$(NCCO), the electron pocket centered at $(\pi,0)$ and the holelike pocket centered at (π,π) of the Brillouin zone successively formed when the Ce concentration was increased from underdoping to optimal doping.[8] Other works, such as on the Nernst and Hall effects, were also reported about the two kinds of charge carriers.[3,9,10] However, these experiments were mainly carried out on NCCO since the high-quality single crystals can be obtained. For the $(La,Ce)_2CuO_4$ system, which is a member of the electron-doped cuprate superconductor family, a few works about the transport properties were obtained from the thin films[12–14] since no T'-phase (the superconducting phase with single CuO_2 plane) single crystal can be grown.

To deeply explore the transport issue of $(La,Ce)_2CuO_4$, especially to distinguish the role of the electrons and holes in the transport (i.e., to understand the evolution of charge carriers in the transport), we carefully measured the Hall resistivity (ρ_{xy}) and longitudinal resistivity (ρ_{xx}) of the optimally doped $(La,Ce)_2CuO_4$ [i.e., the (001)-oriented $La_{1.89}Ce_{0.11}CuO_4$ (LCCO)] thin films when the temperature and magnetic field are varied. We find that both ρ_{xy} and ρ_{xx} are linear in B^2 in the temperature range of 40–50 K, which seems to show an indication of two kinds of charge carriers in LCCO. In addition, in the temperature range of 14–50K, two sign reversals of the Hall resistivity occurs in the normal state with varying T and under certain magnetic fields, which may also give an indication of the existence of two kinds of charge carriers. The sign reversal may be attributed to the competition between τ_e/τ_h (the relaxation time ratio of electrons to holes) and n_e^*/n_h^* (the effective carrier concentration ratio of electrons to holes) when temperature and field change. Here, the effective concentration of electrons and holes, n_e^* and n_h^*, are defined as the number of electrons and holes that directly contribute to the transport. Furthermore, the sign reversal by tuning the field may be originated from the change of cyclotron orbits.

The process of the film growth has been reported in detail in our previous work.[15,16] The thickness of the LCCO thin films is ~120 nm, and the transition temperature T_c is ~24 K. The thin films were patterned into the bridge with 2100 μm (length) $\times$100 μm (width) by photolithography and ion milling techniques. All the measurements were performed by the Quantum Design PPMS-14 equipment. ac source with amplitude of 0.1 mA and frequency of 333 Hz is used. In order to understand the evolution of effective electrons and holes in the normal state, the measuring temperature is selected to be in the range of 14–50 K, because the electrons are responsible for the transport above 50 K, and in the temperature range far lower than T_c, it is difficult to distinguish whether the sign reversal is due to the vortex motion or the evolution of effective electrons and holes. The magnetic field is selected to be high enough (up to 12 T) to drive the LCCO to be in the normal state below T_c. The measurements were performed in two ways: the temperature and magnetic-field (B) dependences of ρ_{xy} and ρ_{xx} with $\mathbf{B}\perp ab$ plane, and the temperature and angular (θ) dependences of ρ_{xy} and ρ_{xx}, with θ the angle between $\mathbf{B}$ and ab plane. [A sketch is shown in the inset of Fig. 1(b)].

In Fig. 1(a), we show the contour map of ρ_{xy} versus B and T for the case of $\mathbf{B}\perp ab$ plane. In order to distinguish the mixed and normal states, the upper critical field B_{c2}, which is determined by the inflexion of ρ_{xx} versus B at different temperatures, is also marked by crosses. It can be clearly seen that the sign of ρ_{xy} is positive in the temperature region of 15–29 K and magnetic fields of 0–5 T, the Hall coefficient anomaly; i.e., the sign reversal of ρ_{xy} due to the dissipation

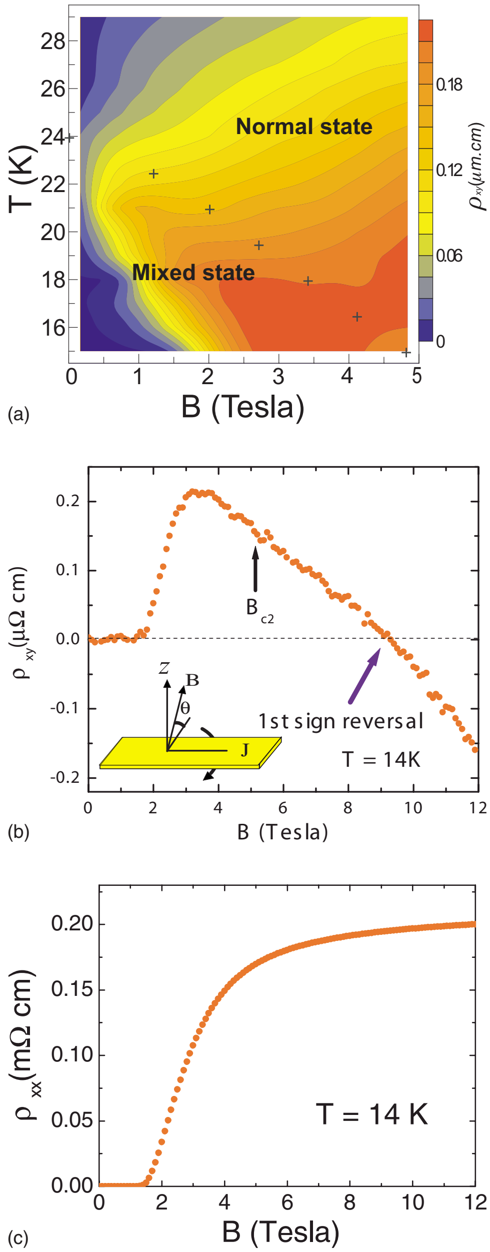

FIG. 1. (Color online) (a) The temperature and magnetic-field dependences of ρ_{xy} under the case of $\mathbf{B}\perp ab$ plane, with B from 0 to 5 T and T from 15 to 29 K. The gray scale represents ρ_{xy} from 0 to 0.24 $\mu\Omega$ cm. The upper critical field B_{c2} is indicated by crosses. (b) ρ_{xy} versus B at 14 K also for $\mathbf{B}\perp ab$ plane. The arrow points to the upper critical field $\sim$5.2 T. It is clear that in the normal state, the sign of ρ_{xy} is reversed from positive to negative with increasing B to $\sim$9.3 T. Inset: a sketch of angular dependence measurement. (c) Corresponding ρ_{xx} versus B at 14 K.

of vortex motion[17] is not observed at the mixed state in this (T,B) region. It is obvious that in the normal state, the all-positive ρ_{xy} indicates that in this (T,B) region the holes play a dominant role in the transport. In detail, the ρ_{xy} increases with increasing B and decreasing T. However, in the lower right part of the contour map, i.e., below 16 K and the field $B\geqslant 4$ T, the ρ_{xy} decreases. It is interesting to know how the ρ_{xy} varies with decreasing temperature and increasing magnetic field. The answer is plotted in Fig. 1(b), which shows the field dependence of ρ_{xy} at 14 K. With increasing B from 0 to 12 T, the ρ_{xy} first keeps a negligible value below 2 T ($<B_{c2}\sim 5.2$ T); this means that no detectable vortex motion contributes to ρ_{xy}. Then, due to the vortex motion, the ρ_{xy} increases, and decreases after reaching a maximum value at $\sim$3.5 T; a sign reversal of ρ_{xy} is clearly observed at the field $\sim$9.3 T in the normal state. This fact obviously indicates that both electrons and holes contribute to the transport in the lower temperature region and that the electrons make more contribution to ρ_{xy} than the holes with increasing magnetic field. The field dependence of ρ_{xx} at 14 K is also shown in Fig. 1(c) and no obvious change can be found when the sign reversal occurs.

To understand the evolution of effective electrons and holes of LCCO thin films in the normal state at a relatively wide temperature region, we focus our measurements on the field angular dependence of ρ_{xy} and ρ_{xx} at $T>T_c$. The $\rho_{xy}(\theta)$ is shown in Fig. 2(a). With increasing T from 25 to 50 K, ρ_{xy} experiences a remarkable change. Under 10 T, from 25 to 30 K, the sign of ρ_{xy} remains positive while the magnitude is reduced by $\sim$60%. With increasing angle, the ρ_{xy} gradually shows a tendency to be negative. When the temperature is increased to 40 K, the sign of ρ_{xy} changes to negative entirely, and the magnitude increases with increasing θ. At 50 K, the magnitude of ρ_{xy} is about two times larger than that at 40 K. Therefore it is obvious that in the temperature region from 25 to 50 K, there is a strong competition between effective holes and electrons. Especially, it is observed that the magnetic field can induce a dramatic change of the effective charge carrier in a small temperature region from 32 to 35 K. At magnetic field of 6 T, the ρ_{xy} of LCCO thin film shows a positive sign from 0° to 90° at 32 K. While in the same temperature range and at magnetic field of 12 T, the $\rho_{xy}(\theta)$ clearly shows a sign reversal from positive to negative at $\theta\sim 51°$. This is another sign reversal of ρ_{xy}, since we met the sign reversal of ρ_{xy} at 14 K in the case of $\mathbf{B}\perp ab$ plane. At 35 K and 12 T, the sign of $\rho_{xy}(\theta)$ becomes entirely negative as shown in Fig. 2(b). It is obvious that the larger field can change the sign of $\rho_{xy}(\theta)$, because a large θ corresponds to a large field along the z axis, $B_z=B\sin\theta$. In order to confirm the role of B_z on the sign and magnitude of ρ_{xy} (ρ_{xx}), we compare the ρ_{xy} (ρ_{xx}) to $|B_z|$ under the cases of $-90°$–$0°$, 0°–90°, and $\theta=90°$ (the case of $\mathbf{B}\perp ab$ plane) at the same temperatures, and find that the same $|B_z|$ has the same ρ_{xy} (ρ_{xx}). That is, the curves of ρ_{xy} (ρ_{xx}) versus $|B_z|$ under these three cases overlap each other well. Typically, as shown in Figs. 2(c) and 2(d), the curves overlap well whenever we change the magnitude or/and the orientation (θ) of $\mathbf{B}$ at 25 K [$\rho_{xy}(|B_z|)$ at 32 K is also shown here]. So we can draw the conclusion that in the present LCCO thin films, the ρ_{xy} and ρ_{xx} are just determined by $|B_z|$, and the role of B_y ($B\cos\theta$) on ρ_{xy} (ρ_{xx}) can be neglected at $T>T_c$. This is reasonable because the LCCO thin films exhibit basically two-dimensional behavior, and the coupling between the CuO_2 layers is very weak,[15] so the charge carriers are mainly in the CuO_2 plane. ρ_{xy} and ρ_{xx} are hardly affected by B_y. On the other hand, for the optimally doped $(La,Ce)_2CuO_4$ thin films, no antiferromagnetic exchange in-

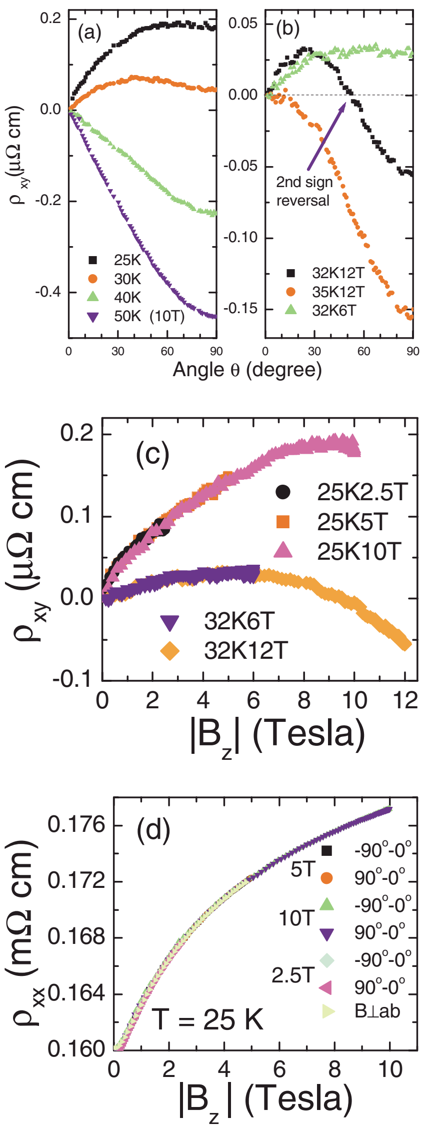

FIG. 2. (Color online) The angular dependence of ρ_{xy} (a) under 10 T: at 25 and 30 K, the ρ_{xy} in positive and gradually shows a negative tendency for increasing θ; at 40 and 50 K, the ρ_{xy} entirely changes to negative, (b) Under 12 T, the sign is negative at 35 K and the sign reversal occurs at 32 K and B_z=9.6 T. In comparison with the case of 6 T at 32 K (triangles), such sign reversal is clearly caused by the magnetic field. The curves of (c) $\rho_{xy}(|B_z|)$ at 25 and 32 K, and (d) $\rho_{xx}(|B_z|)$ at 25 K; they overlap very well under the following from cases of θ: $-90°$ to $0°$, from $0°$ to $90°$, and θ=90° (the case of $\mathbf{B}\perp ab$ plane), which indicates that the ρ_{xy} and ρ_{xx} are dominated by B_z, and the effect of B_y can be neglected.

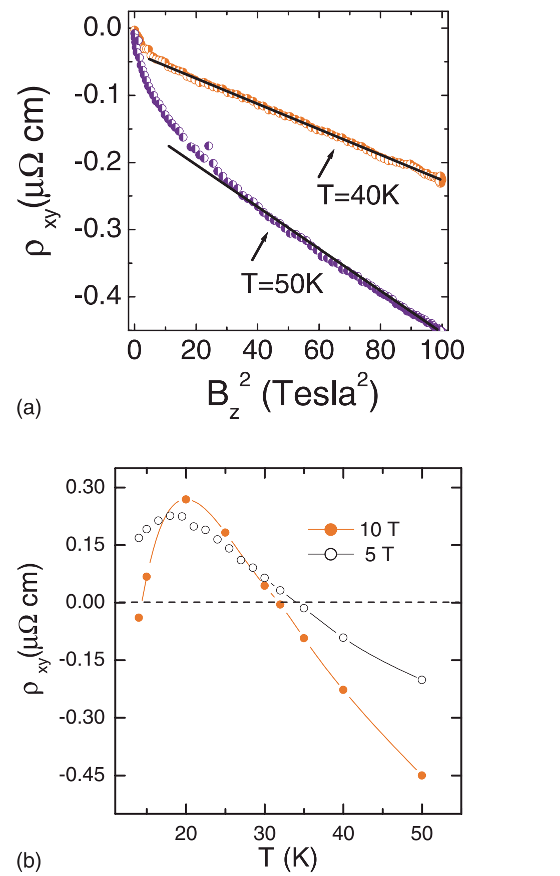

FIG. 3. (Color online) (a) The scaling law of $\rho_{xy}(|B_z|)$ at 40 and 50 K. It is obvious that ρ_{xy} is linear with B_z^2 (the thin lines), and ρ_{xx} is also proportional to B_z^2 (not shown here), so LCCO in this temperature region behaves as a compensated metal, in which the concentrations of electrons and holes are nearly equal. (b) ρ_{xy} versus T for B=5 and 10 T; two sign reversals are clearly indicated.

teraction and spin scattering exist, which is also the reason for the overlapping of $\rho_{xx}(|B_z|)$.[18]

In order to definitely know if both the electrons and holes contribute to the transport in LCCO, we plot the ρ_{xy} versus B_z^2 in Fig. 3(a). It is obvious that when the applied field is larger than a certain value and the temperature is in the range of 40–50 K, the ρ_{xy} shows a linear relationship with B_z^2. ρ_{xx} also shows the same field dependence in this temperature region (not shown here), similar to the ρ_{xx} behavior in $Nd_{1.85}Ce_{0.15}CuO_4$.[11] The behavior of ρ_{xy} and ρ_{xx} proportional to B_z^2 looks like the feature of a compensated metal, in which nearly equal effective concentrations of holes (n_h^*) and electrons (n_e^*) in LCCO is suggested.[19] This is a further evidence that both electrons and holes contribute to the transport in LCCO at lower temperature.[14,20] While at higher temperature (>50 K), only the electrons play a dominant role *in* the transport. Then we may map the evolution of the role of electrons and holes in the transport in the present LCCO thin films. Above 50 K, the electrons are the dominant charge carriers in the transport. From 50 to 40 K, the electrons and holes that contribute to the transport are nearly equal, i.e., $n_h^* \approx n_e^*$, in the larger field region. From 35 K to near T_c, n_h^*/n_e^* likely increases rapidly with decreasing T, and holes are the dominant carriers in the transport, while a large field can enhance the contribution of electrons to Hall resistivity and lead to a sign reversal occuring at 32 K ($\theta \sim 51°$, and $B\sin\theta \sim 9.6$ T). When the temperature is decreased further, the larger field and lower T induce a relatively large contribution of electrons to Hall resistivity again; that is, the sign reversal from positive to negative occurs at 14 K and 9.3 T. In order to conclude the evolution of the role of electrons and holes in transport with temperature, the temperature dependence of ρ_{xy} is plotted in the fixed magnetic fields as shown in Fig. 3(b). It is obvious that the high field ($\sim$10 T) induces two sign reversals of ρ_{xy} at 14 K and $\sim$32 K.

Now, we make a discussion on the possible origin of the sign reversal of ρ_{xy}. Owing to the two kinds of charge carriers, we can express the ρ_{xy} using the two-band model,[21,22]

$$\rho_{xy} = \frac{A_e\gamma_e + A_h\gamma_h}{(A_e + A_h)^2 + (A_e\gamma_e + A_h\gamma_h)^2},$$

with

$$A_i \equiv \sigma_i/(1+\gamma_i^2),$$

where σ_i is the conductance of ith band; the subscripts e and h represent electron and hole, respectively; and $\gamma_i \equiv \omega_i\tau_i$, with ω_i and τ_i the cyclotron frequency and average relaxation time, respectively. Obviously, the sign reversal occurs under the condition $A_e\gamma_e + A_h\gamma_h = 0$ ($\rho_{xy}=0$), i.e.,

$$\frac{n_h^*\tau_h^2\omega_h}{m_h^*(1+\omega_h^2\tau_h^2)} = \frac{n_e^*\tau_e^2\omega_e}{m_e^*(1+\omega_e^2\tau_e^2)}.$$

For $\omega\tau \ll 1$, the above equation can be simplified as $n_h^*\tau_h^2\omega_h/m_h^* = n_e^*\tau_e^2\omega_e/m_e^*$. Note that $\omega_e = eB/m_e^*c$, $\omega_e/\omega_h \propto m_h^*/m_e^*$; if we assume that m_h^*/m_e^* (the ratio of cyclotron masses) is constant when T and B are varied, then the sign reversal should be associated with τ_h/τ_e and n_h^*/n_e^*. If these ratios increase, the sign of ρ_{xy} trends toward positive, or else it trends toward negative. In fact, these two terms may not increase (or decrease) simultaneously, so sign reversal can occur naturally. At 32 K, the sign is positive at lower field and negative at higher field. One possible way to interpret the sign reversal with increasing B is that the cyclotron orbits of charge carriers can be divided into open and closed ones; for the latter, due to the intersection of the Fermi surface and the Brillouin zone, it can behave as either electron- or hole-like orbits,[19] so the sign lies in the closed orbits. At lower field, the relatively lower value of ω_e results in the case that a large number of cyclotron orbits behave as open ones; larger n_h^*/n_e^* makes the sign positive, On the other hand, a high field results in the suppression of the open cyclotron orbits, the ω_e will be increased higher than ω_h, and relatively more open orbits change into the n-type closed orbits; that is, increasing B corresponds to increasing the effective concentration ratio of electrons to holes (n_e^*/n_h^*), and so the sign reversal of ρ_{xy} from positive to negative occurs. If T is decreased, n_h^*/n_e^* increases,[3,14] and the holes become the dominant charge carriers in the transport. We should also note that when decreasing T, τ_h/τ_e is reduced. This is because the scattering in a n-type band is stronger than that in a p-type band at high temperature; when the temperature is reduced, the umklapp processes for a n-type band tends to be "frozen out," so τ_e in a n-type band is increased[22] and τ_h/τ_e is reduced, From $\frac{1}{\tau} = \int\Theta(K,K')\,(1-\cos\eta)dK'/(2\rho)^3$ (η is the scattering angle),[23] we know that decreasing T can reduce the large-angle scattering, so the integral is reduced and τ_e increases. Experimentally, in the temperature region near T_c, the sign reversal of ρ_{xy} from negative to positive

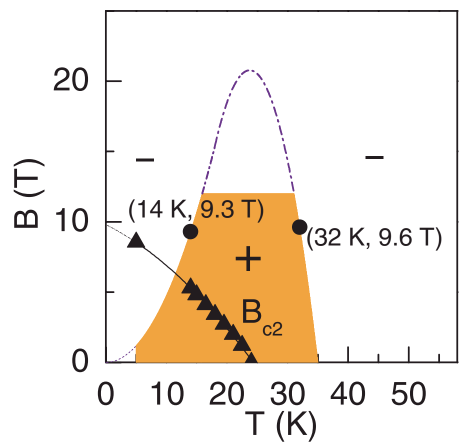

FIG. 4. (Color online) The schematic map of the sign of ρ_{xy} versus B and T (+ and − regions represent positive and negative, respectively), which is obtained by simulation of $\rho_{xy}=0$ with α =0.5 and β=1 (Ref. 25) for low temperature. The circles mark the two sign reversals we observed. It is obvious that through the + region, sign reversal due to the evolution of the two kinds of charge carriers occurs. The upper critical field is also plotted (triangles).

usually occurs, It seems that n_h^*/n_e^* increases more quickly than $(\tau_e/\tau_h)^2$ with decreasing T, so holes become the dominant carriers in the transport in this region. However, when T is decreased further, $(\tau_e/\tau_h)^2$ increases more steeply than n_h^*/n_e^*, then a larger field may cause the sign reversal again (such as the case at 14 K and at 9.3 T). Therefore, we assume $\tau_e/\tau_h \propto 1/T$ [$\tau_e \propto 1/T^2$, (Ref. 2) and $\tau_h \propto 1/T$ (Refs. 2 and 24)], and $n_h^*/n_e^* \propto B^{-\alpha}T^{-\beta}$, with α and β the positive constants in the low-temperature region,[10] Then a schematic map of the sign of ρ_{xy} versus T and B can be made as shown in Fig. 4. By the way, it should be emphasized that we cannot tell which kind(s) of charge carriers is essential to superconductivity merely from the Hall measurements, because both the two kinds of charge carriers contribute to the transport at low T. In order to get a definite decision, further investigations on electron-doped cuprate superconductors are required.[16]

In summary, to reveal the evolution of the role of electrons and holes in the transport in LCCO, we carefully measured the temperature, field, and angular dependences of Hall resistivity and longitudinal resistivity, and find evidence of two kinds of charge carriers ($\rho_{xy}, \rho_{xx} \sim B_z^2$). In the normal state, two sign reversals of the Hall resistivity occurs with varying T for a certain magnetic-field region, which may be attributed to the strong competition between τ_e/τ_h and n_e^*/n_h^* in the temperature range of 14–50 K. The possible reason of sign reversal by tuning the field is attributed to the change of cyclotron orbits of the charge carriers, and the sign reversal against (B,T) is concluded in Fig. 4.

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[25] Y. Ando *et al.* [Y. Ando, Y. Kurita, S. Komiya, S. Ono, and K. Segawa, Phys. Rev. Lett. **92**, 197001 (2004)], have reported that in slightly doped $YBa_2Cu_3O_x$ and $La_{2-y}Sr_yCuO_4$, in-plane resistivity ρ changes as $\sim T^2$. If $\tau_h \propto 1/T$, then simply from $\sigma = ne^2\tau/m^*$ we may get $n_h^* \propto 1/T$. $n_h^*/n_e^* \propto B^{-1/2}$ is obtained from the experimental data, which is still puzzling and not shown here. We mention that though all the assumptions may be simplified, the sign reversal due to changing T and **B** is convinced.