Hall coefficient in the ground state of the charge stripe-ordered La$_{2-x}$Ba$_x$CuO$_4$ single crystals

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Temperature dependence of the Hall coefficient, $R_H$, has been investigated in charge-spin stripe-ordered La-214 high-$T_c$ superconductors. Using the simplest stripe-ordered system of La$_{2-x}$Ba$_x$CuO$_4$, it has been clarified for the first time, to our knowledge, that both the behavior of $R_H$ and its sign exhibit significant dependences on the hole concentration. That is, $R_H$ is zero in the ground state of the charge-spin stripe order at $x = 1/8$, while it is negative in the less-stabilized state of the charge stripe for $x < 1/8$. These are interpreted as being due to the delicate balance of the contributions of the hole-like Fermi surface and the possible electron pocket arising from the formation of the charge-spin stripe order.

The Hall coefficient, $R_H$, in the high-$T_c$ superconducting (SC) cuprates has attracted considerable attention due to its peculiar behavior since the early stage of the high-$T_c$ research. The $R_H$ in the normal state is strongly dependent on temperature, which is unusual in conventional metallic superconductors. The temperature dependence of $R_H$ at high temperatures has been explained based upon the Fermi-liquid model [1] or the two-carrier model [2] taking into account spin fluctuations or charge fluctuations, respectively, but the behavior at low temperatures has not yet been understood.

The experimental results of $R_H$ at low temperatures in the La-214 cuprates such as La$_{2-x}$Ba$_x$CuO$_4$ (LBCO) [3–5] and La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ (LNSCO) [6] around $x = 1/8$ have revealed that $R_H$ markedly decreases with decreasing temperature below the structural-phase-transition temperature between the tetragonal low-temperature (TLT) phase (space group: $P4_{2}mnc$) and the orthorhombic mid-temperature (OMT) phase ($Bmab$), $T_{d2}$. This has been explained as being due to the disappearance of the Hall voltage in the one-dimensional (1D) charge stripe-ordered state [8] stabilized through the structural phase transition [9] or due to the cancellation of the Hall voltage by equal numbers of holes and electrons in the charge domain in the stripe-ordered state [10], although it is still controversial. A gradual decrease in $R_H$ in the normal state with decreasing temperature at low temperatures has been observed in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) with $\delta = 0.15 - 0.40$ (Ref. [3]) and slightly Zn-substituted La$_{2-x}$Sr$_x$Cu$_{1+y}$Zn$_y$O$_{4+y}$ with $x = 0.115$ and 0.15, [10] which has been discussed in relation to the formation of the charge stripe order.

Recently, measurements of $R_H$ in high magnetic fields for underdoped YBCO with the hole concentration per Cu in the CuO$_2$ plane, $p = 0.10 - 0.14$ by LeBoeuf et al. [11] have revealed a sign change of $R_H$ at low temperatures. They have proposed that the negative $R_H$ originates from the formation of an electron pocket through the reconstruction of the Fermi surface caused by the possible formation of the charge stripe order. In YBCO, however, effects of the possible charge stripe order on various physical properties are relatively weak compared with those in the La-214 cuprates, preventing one from understanding explicitly the relation between the electron pocket and the charge stripe order.

In this Letter, $R_H$ in the charge stripe-ordered state is investigated in La-214 cuprates with various values of $p$. It has been found for the first time, to our knowledge, in LBCO with $x = 0.08 - 0.12$ and LNSCO with $x = 0.12$ that the behavior of $R_H$ including the sign at low temperatures exhibits a significant dependence on $p$. That is, $R_H$ for $x = 0.10 - 0.12$ undergoing the phase transition to the TLT phase markedly decreases with decreasing temperature below $T_{d2}$. Moreover, a sign change of $R_H$ is observed at low temperatures for $x = 0.10$ and the absolute value of the negative $R_H$ decreases with increasing $x$, followed by almost zero for $x = 0.12$ where the charge stripe order is completely stabilized. These results indicate that $R_H$ is zero in the ground state of the charge-spin stripe order at $x \sim 1/8$ and that $R_H$ becomes negative in the less-stabilized state of the charge stripe. It appears that there exists a close correlation between the stability of the charge stripe order, the sign of $R_H$ and the topology of the Fermi surface discussed later.

Single crystals of LBCO with $x = 0.08, 0.10, 0.11, 0.12$ and LNSCO with $x = 0.12$ were grown by the traveling-solvent floating-zone method. The detailed procedures are described elsewhere. [12] The composition of each crystal was analyzed by the inductively-coupled-plasma analysis. For LBCO and LNSCO with $x = 0.10 - 0.12$, the charge stripe order is formed at low temperatures below $T_{d2}$ [13, 14] while it is not for LBCO with $x = 0.08$. [14] Both $R_H$ and the ab-plane electrical resistivity, $\rho_{ab}$, were measured by the standard ac six-probe method in magnetic fields parallel to the c-axis up to 9 T, using a commercial apparatus (Quantum Design, PPMS). Both temperature and magnetic-field dependences of the Hall voltage were measured, resulting in good agreement of
values of $R_H$ each other.

Figure 1 shows the temperature dependence of $\rho_{ab}$ in zero field and a magnetic field of 9 T parallel to the $c$-axis normalized by its value at 80 K, $\rho_{ab}^{08K}$, for $La_{2-x}Ba_xCuO_4$ with $x = 0.08 - 0.12$ and $La_{1.6-x}Nd_xSr_xCuO_4$ (LNSCO) with $x = 0.12$. Note for $La_{2-x}Ba_xCuO_4$ with $x = 0.12$ that the descriptions 'A' and 'B' correspond to different batches of the crystal. Arrows indicate the structural-phase-transition temperature between the TLT and OMT phases, $T_{d2}$.

because their reported $\rho_{ab}$ in zero field is metallic even below $T_{d2}$, suddenly drops around 40 K and decreases gradually toward zero with decreasing temperature.

Temperature dependences of $R_H$ and $\rho_{ab}$ in various magnetic fields up to 9 T are displayed in Fig. 2 for LBCO with $x = 0.10$. It is noted that similar temperature dependences of $R_H$ and $\rho_{ab}$ are observed for LBCO with $x = 0.11$ (Ref. [5]). With increasing field, the SC transition curve in $\rho_{ab}$ vs. $T$ exhibits broadening characteristic of the underdoped high-$T_c$ cuprates. The $R_H$ is almost independent of temperature above $T_{d2}$, whereas it suddenly decreases below $T_{d2}$. Moreover, a sign change of $R_H$ is observed below about 26 K and eventually $R_H$ goes to zero at low temperatures. Since temperatures at which $R_H$ and $\rho_{ab}$ become zero are in good agreement with each other, the behavior of $R_H$ going to zero at low temperatures is due to the SC transition. Accordingly, it is concluded that the sign of $R_H$ in the ground state of LBCO with $x = 0.10$ is negative.

Figure 3 shows the $p$-dependent behavior of $R_H$ in 9 T parallel to the $c$-axis. For LBCO with $x = 0.11$, data of $R_H$ obtained from the measurements of the magnetic-field dependence of the Hall voltage at 70 K (above $T_{d2}$), 45 K (a little below $T_{d2}$) and 30 K (at a temperature with negative $R_H$) are also plotted, exhibiting a good agreement with data of $R_H$ obtained from the measurements of the temperature dependence of the Hall voltage in 9 T. At high temperatures around 80 K, $R_H$ is found to be positive and decrease with increasing $x$, as in the case of $La_{2-x}Sr_xCuO_4$ (LSCO) single crystals. For $x = 0.08$, $R_H$ above the onset temperature of the SC transition,

![Graph](image1.png)

**FIG. 1:** (color online) Temperature dependence of the ab-plane electrical resistivity, $\rho_{ab}$, in zero field and a magnetic field of 9 T parallel to the $c$-axis normalized by its value at 80 K, $\rho_{ab}^{08K}$, for $La_{2-x}Ba_xCuO_4$ with $x = 0.08 - 0.12$ and $La_{1.6-x}Nd_xSr_xCuO_4$ (LNSCO) with $x = 0.12$. Note for $La_{2-x}Ba_xCuO_4$ with $x = 0.12$ that the descriptions 'A' and 'B' correspond to different batches of the crystal. Arrows indicate the structural-phase-transition temperature between the TLT and OMT phases, $T_{d2}$.

![Graph](image2.png)

**FIG. 2:** (color online) Temperature dependences of the Hall coefficient, $R_H$, (left axis) and the ab-plane electrical resistivity, $\rho_{ab}$, (right axis) in various magnetic fields parallel to the $c$-axis up to 9 T for $La_{2-x}Ba_xCuO_4$ with $x = 0.10$. The arrow indicates the structural-phase-transition temperature between the TLT and OMT phases, $T_{d2}$.
The marked decrease in $R_H$ below $T_{d2}$ is discussed. The sudden decrease in $R_H$ accompanied by the formation of the charge stripe order has formerly been observed in LBCO \cite{8} and LNSCO \cite{6}. These former results of $R_H$ are compatible with the present ones. A possible origin of the decrease in $R_H$ together with the formation of the charge stripe order has been proposed by Noda \textit{et al.} \cite{6} to be the disappearance of the Hall voltage due to the formation of a 1D charge domain in the stripe-ordered state. From direct numerical calculations including a stripe potential within the $t$-$J$ model, on the other hand, Prelovšek \textit{et al.} \cite{8} have suggested that equal numbers of holes and electrons due to the half occupancy of holes in the charge domain leads to the disappearance of the Hall voltage. For $x = 0.10$ and 0.11 where the half occupancy in the 1D charge domain is guessed to be maintained, \cite{11} however, $R_H$ exhibits negative finite values at low temperatures, as shown in Fig. 3. Therefore, these two ways of thinking are not enough to explain the low-temperature behavior of $R_H$ in the stripe-ordered state.

The sign change of $R_H$ is often observed in the SC fluctuation regime, which is understood to be due to the vortex motion. In this case, it has been suggested that the application of magnetic field tends to suppress the sign change. \cite{17} As shown in Fig. 2, however, the sign change is not suppressed but enhanced by the application of magnetic field, and moreover, $R_H$ is negative even in the normal state above $T_{\text{c}\text{mest}}$ for $x = 0.10$ and 0.11. Therefore, the vortex motion is not the origin of the sign change of $R_H$ in LBCO with $x = 0.10$ and 0.11. Simply thinking, the sign of $R_H$ is sensitive to the subtle curvature of the Fermi surface. As mentioned before, recently, LeBoeuf \textit{et al.} \cite{11} have found a pronounced sign change of $R_H$ at low temperatures in strong magnetic fields in underdoped YBCO with $p = 0.10 - 0.14$. Since quantum oscillations have been observed in YBCO with $p = 0.10$, \cite{18} they have suggested that the negative $R_H$ is a product of the formation of an electron pocket through the Fermi-surface reconstruction caused by the possible formation of the charge stripe order. A similar behavior of $R_H$ has formerly been found in 2H-TaSe$_2$ where both a strong decrease and a sign change of $R_H$ are observed accompanied by the transition to the charge-density-wave (CDW) state. \cite{17} These have theoretically been explained in terms of the Fermi-surface reconstruction due to the opening of the CDW gap. \cite{20} Accordingly, the sign change of $R_H$ at low temperatures in LBCO with $x = 0.10$ and 0.11 is possibly explained as being due to the creation of an electron pocket on the Fermi surface caused by the formation of the charge stripe order.

Then, why does the behavior of $R_H$ at low temperatures depend on $p$ significantly? Supposed that the Fermi surface in the underdoped cuprates is reconstructed through the formation of a commensurate antiferromagnetic (AF) order or a $d$-density-wave (dDW) order. \cite{21} both hole pockets and an electron pocket are cre-

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{(color online) Temperature dependence of the Hall coefficient, $R_H$, in 9 T parallel to the $c$-axis for La$_{2-x}$Ba$_x$CuO$_4$ (LBCO) with $x = 0.08 - 0.12$ and La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ (LNSCO) with $x = 0.12$. Note for LBCO with $x = 0.12$ that the descriptions 'A' and 'B' correspond to different batches of the crystal. Solid arrows indicate the structural-phase-transition temperature between the TLT and OMT phases, $T_{d2}$. Open arrows indicate the onset temperature of the SC transition, $T_{\text{c} \text{mest}}$, in 9 T estimated from $\rho_{ab}$ shown in Fig. 1. Open diamonds show data of $R_H$ obtained from the measurements of the magnetic-field dependence of the Hall voltage for LBCO with $x = 0.11$. The inset is a schematic drawing of one possible reconstruction of the Fermi surface through the formation of a commensurate antiferromagnetic order or a $d$-density-wave order \cite{21} for LBCO with $x = 0.10$ and 0.12. $T_{\text{c} \text{mest}}$, estimated from $\rho_{ab}$ in 9 T shown in Fig. 1, is almost independent of temperature, whereas it gradually decreases with decreasing temperature below $T_{\text{c} \text{mest}}$ due to the SC transition. For $x = 0.11$ and 0.12, on the other hand, $R_H$ markedly decreases below $T_{d2}$ as well as for $x = 0.10$ shown in Fig. 2. Since $T_{\text{c} \text{mest}}$ is far below $T_{d2}$, the marked decrease in $R_H$ is irrespective of the SC transition. Comparing the data of $x = 0.10$ with those of $x = 0.11$, the extrapolation of the data above $T_{\text{c} \text{mest}}$ to zero temperature suggests that the sign change of $R_H$ is more marked for $x = 0.10$ than for 0.11, as shown by broken lines in Fig. 3. For LBCO and LNSCO with $x = 0.12$, on the other hand, $R_H$ becomes almost zero at low temperatures. These results are summarized as follows. For the samples in the TLT phase and therefore in the charge stripe-ordered state, $R_H$ markedly decreases with decreasing temperature below $T_{d2}$. Moreover, the sign change of $R_H$ at low temperatures exhibits a significant dependence on $p$.}
\end{figure}
ated on the Fermi surface, located around \((\pm \pi/2, \pm \pi/2)\) and \((\pm \pi, 0)\), \((0, \pm \pi)\) in the reciprocal lattice space, respectively, which is schematically shown in the inset of Fig. 3. Actually, recent angle-resolved photoemission experiments have revealed a pocket around \((\pi/2, \pi/2)\).\(^{22,23}\) Considering the \(p\)-dependent change of the Fermi-surface topology,\(^{24}\) the possible electron pocket around \((\pm \pi, 0)\), \((0, \pm \pi)\) tends to shrink with increasing \(p\), as shown in the inset of Fig. 3. This is naturally suggestive of the weakening of the electron aspect and the development of the hole aspect on \(R_H\) with increasing \(p\). This picture is consistent with the present results that the sign change of \(R_H\) gradually weakens and \(R_H\) at low temperatures becomes zero with increasing \(x\) from \(x = 0.10\) to 0.12. It is noted that \(R_H\) is positive at low temperatures for LNSCO with \(x = 0.15\),\(^{6}\) which is also consistent with this picture. Accordingly, the \(p\)-dependent change of \(R_H\) in the ground state is able to be understood by the delicate balance of the contributions of hole and electron pockets.

Millis \textit{et al.}\(^{23}\) and Lin \textit{et al.}\(^{26}\) have theoretically investigated the Fermi-surface reconstruction within the tight-binding model including the charge and spin stripe potential. According to their calculations, rather complicated arrangement of electron pockets and hole pockets is reconstructed and the development of the spin stripe correlation makes \(R_H\) negative, while the development of the charge stripe correlation makes \(R_H\) positive. That is, in the charge-spin stripe-ordered state, the sign of \(R_H\) depends on the delicate balance of the developments of the charge and spin stripe order. Based upon their calculations, it follows that \(R_H\) becomes zero due to complete developments of both charge and spin stripe order at \(x = 1/8\) and that \(R_H\) becomes negative due to the incomplete development of the charge stripe order for \(x < 1/8\).\(^{14}\) Actually, the sign change of \(R_H\) has been observed in YBCO with \(p = 0.10 - 0.14\) (Ref.\(^{11}\)) and LSCO with \(x = 0.12\) (Ref.\(^{27}\)) where the spin stripe correlation is developed but the charge stripe order is not. Accordingly, it may be the case that \(R_H\) becomes zero due to the stabilization of the charge-spin stripe order at \(x = 1/8\), while it becomes negative to the instability of the charge stripe order for \(x < 1/8\).

Finally, we comment on the value of \(R_H\) in the ground state of LBCO. For \(x = 0.10\), the extrapolated value of \(R_H\) to zero temperature is estimated from Fig. 3 to be \(-0.00375\) cm\(^3\)/C. Thus, based on the simple one-carrier model, the carrier density \(n_{\text{Hall}}\) is calculated to be \(n_{\text{Hall}} = -V_{\text{cell}}/eR_H/2 = 0.0795\) per Cu in the CuO\(_2\) plane, where \(V_{\text{cell}}\) is the volume of the unit cell in the notation of the tetragonal at high temperature (14/mm). For YBCO with \(p = 0.10\),\(^{11}\) on the other hand, it has been reported that \(n_{\text{Hall}} = 0.0145\) per Cu in the CuO\(_2\) plane. This value is comparable to the deduced value of 0.019 per Cu in the CuO\(_2\) plane from the quantum-oscillation experiments.\(^{18}\) Assuming the negative value of \(R_H\) is originated from a possible pocket on the Fermi surface, a simple comparison of \(n_{\text{Hall}}\) between LBCO and YBCO results in a larger pocket in LBCO than in YBCO. This appears to be inconsistent with that expected from the experimentally observed Fermi-surface topology,\(^{24,25}\) because the reconstructed Fermi surface due to the commensurate AF order or the draping order is expected to produce a larger electron pocket around \((\pm \pi, 0)\), \((0, \pm \pi)\) in YBCO than in LBCO. Accordingly, it is hard to make quantitative discussion on the negative value of \(R_H\) in the ground state using the simple one-carrier model.

In conclusion, \(R_H\) in the stripe-ordered LBCO and LNSCO in the TLT phase markedly decreases due to the formation of the charge-spin stripe order. In the ground state, on the other hand, \(R_H\) is zero in the completely ordered charge-spin stripe state at \(x = 1/8\), while it is negative in the less-stabilized state of the charge stripe for \(x < 1/8\). The \(p\)-dependent behavior of \(R_H\) including its sign is interpreted as being due to the delicate balance of the contributions of the hole-like Fermi surface and the possible electron pocket arising from the formation of the charge-spin stripe order.

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