Hall effect in the metallic antiferromagnet Na_xCoO_2 ($0.72 \leqslant x \leqslant 0.90$)

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We have investigated the magnetic and magnetotransport properties of Na_xCoO_2 single crystals in the heavily doped region ($0.72 \leqslant x \leqslant 0.90$). Both the Hall coefficient (R_H) and the susceptibility (χ) exhibit strong temperature dependence in the antiferromagnetic (AFM) as well as in the paramagnetic (PM) states. As the AFM ordering sets in, below $T_N = 21$ K, R_H decreases sharply with T down to ~10 K and then increases rapidly. The temperature and field dependence of R_H indicates the emergence of a weak ferromagneticlike phase below 10 K. In the AFM state, R_H scales linearly with χ. The observed T dependence of R_H in the PM state has been discussed in light of the theoretical model proposed for strongly correlated electrons in a triangular lattice.

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I. INTRODUCTION

The layered sodium cobaltite Na_xCoO_2 has attracted considerable experimental and theoretical attention for its anomalous physical properties and rich phase diagram.[1–17] The initial interest in this compound was to understand the origin of the unusually large thermopower with good metallic conductivity.[2,3] Subsequent studies have revealed that superconductivity and many other complex physical phenomena arise due to the strong electronic correlation, the coupling of magnetic states with electronic states near the Fermi level, and the specific crystal structure of the system.[3,4,7–14] The interplay between the magnetic fluctuations and the superconductivity is one of the central issues not only in this system, but also in cuprates, heavy fermions, and iron pnictides. The fundamental building blocks related to the charge conduction and magnetic properties in Na_xCoO_2 are the CoO_2 layers. The two-dimensional metallic CoO_2 planes are electronically separated by the insulating layers of Na ions, and the Co ions in each CoO_2 plane are arranged in a triangular lattice. Because of the triangular geometry, the undoped compound ($x = 0$) with spin-1/2 Co^{4+} is speculated to be a spin-frustrated Mott insulator. With Na doping, charge carriers are introduced, and the spin-1/2 Co^{4+} changes into spinless Co^{3+}. Thus, Na doping enhances conductivity and relaxes magnetic frustration as a result of which a long-range antiferromagnetic (AFM) spin-density-wave (SDW) ordering occurs for $x > 0.70$ with $T_N \sim 21$ K. In the AFM state, the Co spins are ferromagnetically aligned in the plane and are stacked antiferromagnetically along the c axis. Unlike other layered materials, the intralayer and interlayer magnetic interactions in Na_xCoO_2 are of comparable strength, suggesting an essentially three-dimensional nature of the magnetism.[18]

The magnetic, electronic, and thermal properties reveal that several unusual phenomena emerge with the increase in Na content in the heavily doped region ($x > 0.70$).[12–14] The main findings are the strong enhancement of the electronic density of states and spin fluctuations in a narrow range of x close to 0.70 and the opening of a gap in the Fermi surface due to the SDW ordering above $x = 0.70$.[13,14] For $x \geqslant 0.75$, the spin fluctuations get suppressed, and a long-range ferromagnetic-(FM)-like ordering sets in around 8 K. Besides these intriguing phenomena, the breaking of charge homogeneity of the Co ions that stabilizes a charge-ordering-like state and the intricate high-temperature Na ordering are the other remarkable aspects in this doping region.[9,15] Both experimental and theoretical studies reveal that all these phenomena deeply modify the low-energy electronic structure of the system.[8–14] As a consequence, one expects unusual physical properties and a rich interplay of spin and charge degrees of freedom in the heavily doped region.

The Hall effect is one of the most widely used measurements to characterize the low-energy electronic state of a system. Often, the Hall effect in strongly correlated materials is very different from that in a simple metal. In Na_xCoO_2, the temperature and doping (x) dependence of the Hall coefficient (R_H) has not been studied systematically in the heavily doped region where the electronic correlation is believed to be strong and a long-range magnetic ordering appears at low temperatures. R_H is extremely sensitive to the magnetic ground state of the system also. In this paper, we show that T and x dependences of R_H in the heavily doped region (0.72–0.90) are quite different from those reported for $x \leqslant 0.71$.[4,11] Both the weak T dependence of R_H below ~85 K and the strong T-linear behavior at high temperatures are consistent with the theoretical prediction for a triangular lattice.[10] In the AFM state, below $T_m \simeq 10$ K, R_H increases rapidly with the decrease in T and exhibits a nonlinear H dependence. To the best of our knowledge, there is no experimental evidence on the effect of this low-temperature transition on any transport properties.

II. EXPERIMENTAL DETAILS

Single crystals of nominal compositions Na_xCoO_2 ($0.72 \leqslant x \leqslant 0.90$) were grown by floating-zone technique using an image furnace. The growths were carried out in an oxygen atmosphere with a typical growth rate of 4 mm/h. X-ray powder diffraction shows that all samples are single phases. The chemical composition of the crystal pieces used in the present paper was thoroughly analyzed by energy dispersive x ray (Quanta 200 FEG, FEI). For the determination of the Na:Co ratio, we scanned at several points and found that the variation in Na content is within 2% of the nominal composition. This

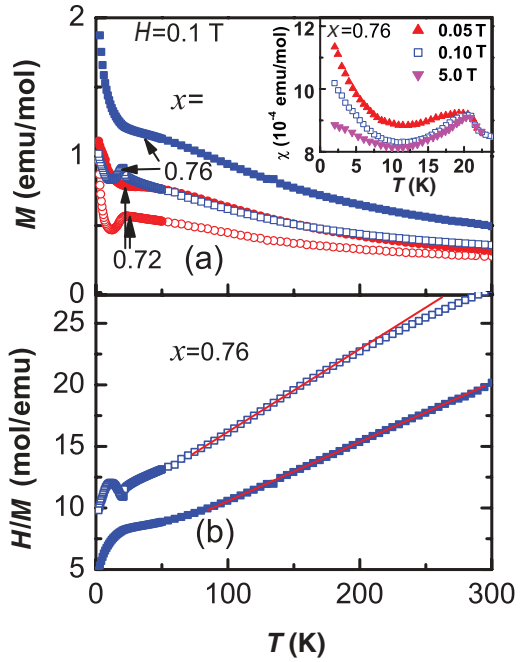

FIG. 1. (Color online) (a) Temperature dependence of magnetization for an applied field $H = 0.1$ T parallel (open symbols) and perpendicular (closed symbols) to the c axis of Na_xCoO_2 single crystals with $x = 0.72$ and 0.76. The inset shows the behavior of $\chi_{\|c}(T)$ below T_N for different H's for the $x = 0.76$ sample. (b) H/M versus T plot for the $x = 0.76$ crystal for $H = 0.1$ T. The solid line shows the linear behavior of H/M.

confirms that these samples are chemically homogeneous. The cleaved crystal was cut into a rectangular shape of the typical size of 3 mm × 2 mm × 0.2 mm. The Hall measurements were performed using a standard four-probe contact with H parallel to the c axis. The Hall voltage was measured at fixed T by slowly sweeping H from −8 to 8 T, and the Hall coefficient (R_H) was calculated using the relation $R_H = \rho_H/H$, where ρ_H is the Hall resistivity. In this method, one can eliminate the longitudinal offset voltage due to a small misalignment of the Hall contacts and can determine H dependence of R_H. Zero-field-cooled magnetization (M) with H parallel ($M_{\|c}$) and perpendicular ($M_{\perp c}$) to the c axis has been measured using a commercial superconducting quantum interference device magnetometer (Quantum Design) with fields up to 5 T.

III. RESULTS AND DISCUSSION

The representative plots of $M(T)$ for $x = 0.72$ and 0.76 samples are shown in Fig. 1(a). M exhibits a strong anisotropy with $M_{\perp c} > M_{\|c}$ over the whole range of T. Upon cooling, the anisotropy increases, and a weak downward curvature appears below 85 K in both $M_{\perp c}$ and $M_{\|c}$. With further lowering of T, the system undergoes a paramagnetic (PM) to AFM transition at $T_N \simeq 21$ K. Below T_N, $M_{\perp c}$ continues to increase rather sharply, whereas, $M_{\|c}$ initially decreases rapidly and then increases for $T < 10$ K. $M_{\perp c}$ does not show any visible change around 10 K, but a weak anomaly is observed in $dM_{\perp c}/dT$. The inset of Fig. 1(a) shows $\chi_{\|c}(T)$ $[=M_{\|c}(T)/H]$ for different H's in the magnetically ordered state. It is clear from the figure that $\chi_{\|c}$ decreases

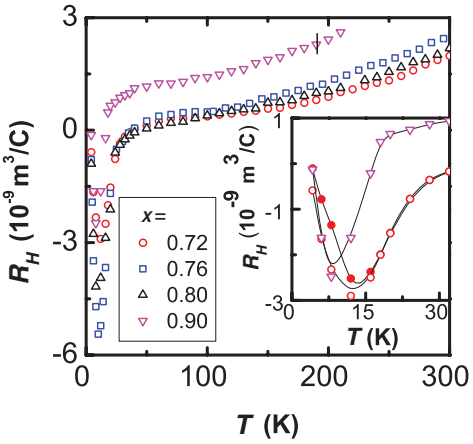

FIG. 2. (Color online) Temperature dependence of the Hall coefficient (R_H) for H (=8 T) parallel to the c axis of Na_xCoO_2 crystals with $x = 0.72$, 0.76, 0.80, and 0.90. The inset shows the enlarged view of $R_H(T)$ for $x = 0.72$ (• for $H = 2$ T and ○ for $H = 8$ T) and 0.90 (▽ for $H = 8$ T) samples below T_N.

systematically with increasing H, but the decrease is faster below 10 K. In order to elucidate the nature of the magnetic ground state, $M(H)$ has also been measured (not shown). Both $M_{\perp c}(H)$ and $M_{\|c}(H)$ are linear above T_N. Below T_N, although $M_{\perp c}(H)$ is linear, $M_{\|c}(H)$ develops a weak hysteresis and nonlinear H dependence at low fields similar to that reported earlier.[6,7] These H and T dependences of $M_{\|c}$ indicate a weak FM-like character. However, the estimated value of spontaneous moment ($\sim 10^{-4}\mu_B$/Co) in the so-called FM state is unusually small compared to a typical FM. Qualitatively, similar behavior has been observed for other compositions. For a better understanding of the PM state, we have plotted the inverse of dc susceptibility χ^{-1} $(=H/M)$ as a function of T for the $x = 0.76$ sample [Fig. 1(b)]. In the PM state, $H/M_{\perp c}$ is almost linear above 85 K, but $H/M_{\|c}$ shows a downward curvature above 180 K, indicating a deviation from the simple Curie-Weiss (CW) behavior. Although $H/M_{\|c}$ starts to deviate from linearity above 180 K, a T-linear dependence is observed in the region 85 K $< T <$ 180 K. It has been suggested that this deviation is due to the presence of a T-independent term in susceptibility.[7] However, Schulze *et al.* have shown that, apart from the high-temperature Na ordering at ∼285 K, there is a further Na rearrangement occurring at around 200 K, and the Coulomb potential due to this Na^+ rearrangement strongly affects the magnetic properties of the sample.[13] In particular, the magnetic transition around 10 K is extremely sensitive to this phenomenon. At present, it is not clear whether the departure in χ is due to the Na rearrangement or presence of a T-independent positive term.

We now turn to the Hall effect results on Na_xCoO_2. R_H with H (=8 T) along the c axis is plotted as a function of T in Fig. 2 for $x = 0.72$, 0.76, 0.80, and 0.90. For all compositions, R_H shows a nonmonotonic T dependence. R_H decreases with T down to T_N and changes its sign around T_N. On further cooling below T_N, a sharp decrease occurs in the interval $T_N > T > T_m$ followed by an abrupt increase in crossing T_m. The inset of Fig. 2 displays $R_H(T)$ for the $x = 0.72$ and 0.90 samples below T_N. For $x = 0.72$, we have plotted $R_H(T)$ for two different fields. The figure shows that the sharp variations

in R_H at T_N and T_m are less sensitive to H. The magnitude of R_H at T_m increases rapidly by a factor of 2 as x increases from 0.72 to 0.76 and then decreases with a further increase in x. The systematic decrease in magnitude of R_H at T_m for $x > 0.76$ and the shift in T_m towards lower T for $x = 0.90$ indicate that the FM interaction gets weakened possibly due to the reduction in the Co^{4+} ion. Closer inspection reveals that several anomalies observed in $M(T)$ are also reflected in $R_H(T)$. Other than the sharp changes at T_N and T_m, a change in slope near 85 K is also common in both $\chi_{\|c}(T)$ and $R_H(T)$. On the contrary, we observe an anomaly in the resistivity only at T_N. This suggests that the transverse resistivity is very sensitive to the magnetic state of the sample.

In the heavily doped region, R_H is large, negative, and shows strong T dependence below T_N. In $Na_{0.5}CoO_2$ too, R_H is large and negative below 75 K and exhibits a minimum around 30 K.[4] However, in this sample, the large R_H is due to the suppression of the charge carrier at the charge-ordering/AFM transition. Also, for $Na_{0.5}CoO_2$, $R_H \to 0$ as $T \to 0$ due to the particle-hole symmetric state. Although we are unable to deduce the value of R_H accurately for $T \to 0$, the extrapolation of the low-temperature data reveals a nonzero positive value of R_H at $T = 0$ for all samples. The nature of $R_H(T)$ for the present samples may be compared with that reported for the metallic $x \leqslant 0.71$ samples.[4,5,19] Comparing our results with those reported earlier, we find that R_H exhibits much sharper variations at T_N and T_m for the present $x \geqslant 0.72$ samples.[4,5,11,19] This difference is possibly due to the lower Na content in the earlier samples. The detailed magnetic, thermal, and muon spin rotation studies reveal that the bulk AFM phase below T_N and the low-temperature phase below T_m appear for $x > 0.70$.[13] This suggests that the amount of ordered magnetic phases present in earlier samples is small and, hence, the reduction in sharpness at T_N and the absence of anomaly below 10 K in $R_H(T)$.[4,11] In order to shade some more light, we have investigated the H dependence of ρ_H both above and below T_N for $x = 0.72$–0.90 samples. Here, we have shown the plots for $x = 0.72$, 0.76, and 0.80 only [Figs. 3(a)–3(c)]. For all compositions, we observe that ρ_H is linear in H above T_N, but it is large and nonlinear in H below T_N. From the figures, we see that $\rho_H(H)$ is almost linear for $12\ \text{K} < T < T_N$. The nonlinearity in $\rho_H(H)$ appears mainly below 12 K and increases with decreasing T. The nonlinear behavior of $\rho_H(H)$ is stronger for the $x = 0.72$ sample. For the $x = 0.90$ sample, we observe that the qualitative behavior of $\rho_H(H)$ is similar to that for $x = 0.76$ and 0.80 samples. The observed nonlinearity and sign change of ρ_H cannot be explained by taking into account two types of carriers with different mobilities because both phenomena occur either below or in the vicinity of T_N. Also, the sign change is accompanied by a sharp drop in $\rho_H(T)$. Although the sign change for the $x = 0.68$ sample occurs smoothly as expected in the presence of two different types of carrier, the nonlinearity in $\rho_H(H)$ appears only below T_N.[5] In ferromagnets, however, $\rho_H(H)$ is nonlinear below T_C and shows an abrupt sign change in the vicinity of T_C. Furthermore, $R_H(T)$ mimics the low field $M(T)$ below T_N. All these facts support the magnetic origin for the nonlinear behavior of $R_H(H)$ below T_N. The weak anomaly at around 7.5 T in $\rho_H(H)$ curves for the $x = 0.80$ sample is due to the metamagnetic transition. Similar to an earlier report,[16] we have also observed a clear signature of metamagnetic transition in magnetoresistance.

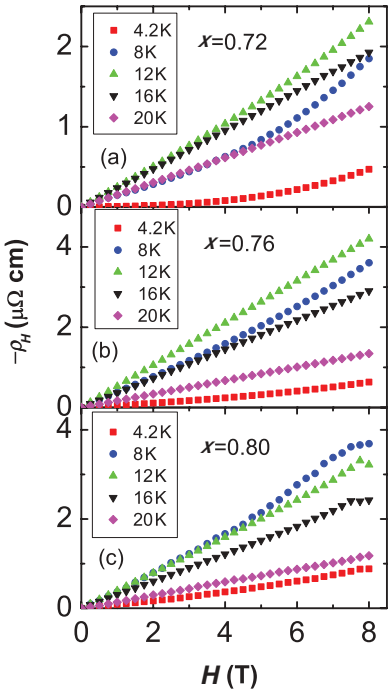

FIG. 3. (Color online) Hall resistivity ρ_H as a function of field with H parallel to the c axis of Na_xCoO_2 crystals for (a) $x = 0.72$, (b) 0.76, and (c) 0.80.

In magnetic materials, often the strong T dependence and sign change of R_H in the vicinity of magnetic transition arise due to the asymmetric scattering of the charge carrier by the local moments.[20–23] To interpret these results, we use the well-known phenomenological expression for the Hall resistivity: $\rho_H = R_o B + R_s M_s$, where R_o is the normal Hall coefficient, R_s is the coefficient of anomalous Hall effect, B is the applied magnetic induction, and M_s is either the spontaneous magnetization or the field-induced magnetization.[21,22] For a simple nonmagnetic metal, the second term is zero, but for a FM metal, this term can be very large below T_C. For T above T_N or T_C, although there is no spontaneous moment contribution to R_H, the PM moment induced by the applied field is important. In the PM state, M_s should be replaced by χH, and one obtains $R_H = R_o + \chi(R_o + R_s)$. So, we expect a linear relation between R_H and χ in the PM state of a magnetic material. This behavior has been experimentally verified for several systems.[21–23] For Na_xCoO_2, the above relation cannot be satisfied because χ decreases while R_H increases with the increase in T above T_N. Usually, M increases linearly with H in the AFM state. Also, in such cases, one expects a linear relation between R_H and χ below T_N. In Fig. 4(a), we have plotted R_H as a function of $\chi_{\|c}$ below T_N for $x = 0.72$ and 0.76 samples. We have plotted R_H versus $\chi_{\|c}$ for two different fields $H = 0.10$ and 5.0 T. For both samples, R_H increases approximately linearly with $\chi_{\|c}$ in the AFM state. However, the data below 8 K do not fall on the line, and their values are slightly larger. This deviation is related to the FM nature of the magnetic state below T_m. Surprisingly enough, we observe an unusual dependence between R_H and $\chi_{\|c}$; instead

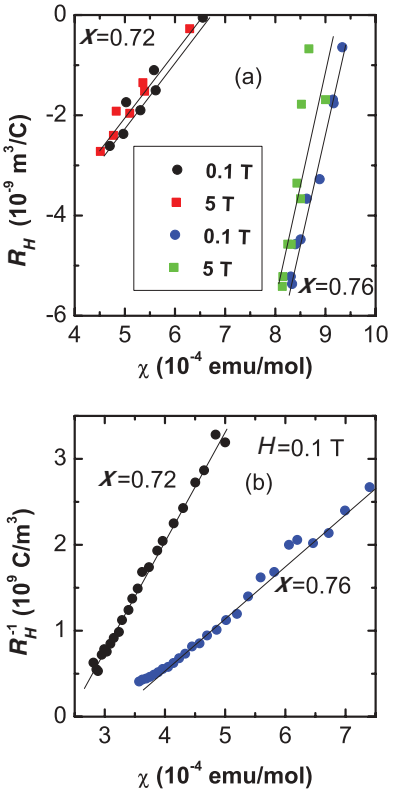

FIG. 4. (Color online) R_H for H parallel to the c axis versus $\chi_{\|c}$ $(=M/H)$ plot for $x = 0.72$ and 0.76 samples: (a) below T_N and (b) in the PM state above 50 K. The different magnetic fields at which both R_H and $\chi_{\|c}$ have been measured are indicated in the figures.

of R_H, R_H^{-1} depends linearly on $\chi_{\|c}$ above ~50 K in the PM state [Fig. 4(b)]. For $x = 0.76$, the deviation from linearity below a critical value of $\chi_{\|c}$ is due to the departure of $\chi_{\|c}(T)$ from the CW law above 180 K. The R_H-$\chi_{\|c}$ plots for $x = 0.80$ and 0.90 samples (not shown) are qualitatively similar to that for $x = 0.72$ and 0.76 samples.

The Hall effect in strongly correlated systems is unusual and, in most cases, the semiclassical expression $R_H = 1/ne$ does not hold well. R_H is rather sensitive to different physical parameters and the crystal geometry of the system. Both cuprates and sodium cobaltites have layered structures, but the nature of T dependence of R_H in the two cases is very different; R_H decreases with the increase in T in the former, whereas, it is just the opposite in the latter. The underlying triangular lattice is believed to play a significant role on the T dependence of R_H in Na_xCoO_2. Shastry *et al.* have shown that, in a triangular lattice, R_H increases linearly with T above the Fermi temperature (T_F), whereas, for $T < T_F$, R_H is less sensitive to T as in the case of a Fermi liquid.[10] The predicted linear behavior is consistent with the reported Hall data for single crystals with $x = 0.68$.[4] However, the value of dR_H/dT estimated by Shastry *et al.* comes out to be a factor of 4 smaller than the experimental one. They speculated that a nearby concentration might reveal a smaller dR_H/dT to be consistent with the experimental value of the hopping-matrix element $t/k_B \sim 100$ K. In this regard, it is useful to compare our Hall data with the theoretical predictions as we have measured the Hall coefficient of the Na_xCoO_2 single crystal for several compositions above $x = 0.68$. Indeed, the approximate high-temperature slope of $R_H(T)$ for our samples is a factor of ~4 smaller than that for the $x = 0.68$ crystal. Thus, the slope of $R_H(T)$ for $x > 0.68$ samples is consistent with the theoretical prediction. The nature of the $R_H(T)$ curve at low T can also be explained using this model. In our samples, R_H is almost T independent and positive for 50 K $< T <$ 100 K, which is in line with the prediction by Shastry *et al.*[10] In this temperature region, we deduce carrier (hole) density 1.5–3 $\times 10^{28}$ m^{-3} for $0.72 \leqslant x \leqslant 0.90$. Also, the extrapolation of $R_H(T)$ data to $T = 0$ suggests that, had there been no AFM ordering for $x \geqslant 0.72$ samples, $R_H(T)$ would tend to saturate at a small positive value at low temperatures as predicted theoretically.

IV. CONCLUSIONS

In conclusion, magnetic and magnetotransport properties of Na_xCoO_2 $(0.72 \leqslant x \leqslant 0.90)$ single crystals with H along the c axis have been studied. R_H shows strong T dependence in the AFM state and, at high temperatures, above 100 K in the PM state. R_H is almost T independent in the region 50 K $< x <$ 100 K. In the AFM state, R_H scales linearly with χ. Both $M(H,T)$ and $R_H(H,T)$ data indicate the emergence of a FM-like magnetic phase below 10 K. The observed T dependence of R_H above T_N is consistent with the theoretically calculated R_H for the triangular lattice in the strong correlation limit. The excellent agreement between theory and experiment is possibly due to the enhancement of electronic correlation above $x \simeq 0.70$.

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