Magnetic percolation effect on the spontaneous Hall resistivity and magnetoresistance of La$_{1-x}$A$_x$CoO$_3$ (A = Ca, Sr; 0.1 ≤ $x$ ≤ 0.5)

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Abstract

The Hall resistivity and magnetoresistance of La$_{1-x}$A$_x$CoO$_3$ (A = Ca, Sr) are investigated. The spontaneous Hall coefficient reaches maximum near the Curie temperature for each doping level, and achieves the largest magnitude near the magnetic percolation threshold. The physical significance of these results are discussed.

The Hall resistivity $\rho_{xy}$ of a metallic system with localized magnetic moments is generally given by $\rho_{xy} = R_0 B + R_s (\mu_0 M)$, where $R_0 = 1/(ne)$ is the Hall coefficient for a conducting carrier density $n$ and charge $e$, $B$ is the magnetic induction, and $R_s$ the anomalous Hall coefficient associated with the magnetization $M$ of a sample. Conventional theory attributes a finite $R_s$ to asymmetric spin-orbit scattering of carriers, and predicts the relations between $\rho_{xy}$ and the longitudinal resistivity ($\rho_{xx}$) as either ($\rho_{xy} \propto \rho_{xx}$) for the skew scattering mechanism [1], or ($\rho_{xy} \propto \rho_{xx}^2$) for the side-jump mechanism [2]. However, our recent studies of ferromagnetic cobaltites La$_{1-x}$Ca$_x$CoO$_3$ (0.2 ≤ $x$ ≤ 0.5) [3] reveal novel properties and a record value of $R_s (\approx 1.4 \times 10^6$ m$^2$/C) in La$_{0.8}$Ca$_{0.2}$CoO$_3$ that cannot be explained by conventional theory. We have attributed these results to the existence of multiple spin configurations of trivalent Co-ions, (Co$^{3+}$: $t_{2g}^3 e_{2g}^1$, $S = 2$; Co$^{III}$: $t_{2g}^6 e_{2g}^0$, $S = 0$), strong spin fluctuations near $T_{Curie}$, and magnetic percolating effect as a function of temperature ($T$) and doping level $x$. In this work, we extend our studies to La$_{1-x}$Sr$_x$CoO$_3$ ($x = 0.2, 0.5$), and compare the properties of Sr-doped with Ca-doped systems, and also with ferromagnetic La$_{1-x}$A$_x$MnO$_3$.

The substitution of divalent Ca or Sr in LuCoO$_3$ results in tetravalent Co$^{4+}$ ($t_{2g}^2 e_{2g}^3$) ions, which stabilizes the high-spin Co$^{3+}$ ions, and ferromagnetism is established for $x > 0.18$ [3]. For a given $x$, the fraction of high-spin Co$^{3+}$ among the trivalent Co-ions increases with $T$, and reaches $\sim 50\%$ at $T \approx 110$ K. Interestingly, $T_{Curie} \approx 110$ K in La$_{0.8}$Ca$_{0.2}$CoO$_3$, and $R_s (T \approx T_{Curie})$ reaches a record value among all known stochiometric ferromagnetic materials [3]. The enhancement of $R_s$ near the magnetic percolation threshold ($x = 0.2$) may be understood as follows: The spontaneous Hall effect is proportional to the spin-orbit coupling strength $\lambda_{so}$, and $\lambda_{so} \sim \langle |k \times \sigma| \cdot \nabla V_c \rangle$, where $V_c$ is the crystalline potential. Ferromag-

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magnetic cobaltites consist of high-spin hole-rich clusters embedded in a background of low-spin hole-poor matrix. Therefore, a maximum magnitude in $\nabla V_c$ is expected near the magnetic percolation threshold, yielding enhanced spin-orbit scattering and large $R_s$. This argument is consistent with the experimental observation in ferromagnetic $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$, as summarized in Figures 1(a)-(b). Similar behavior is also observed in ferromagnetic $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$, as shown in Figures 1(c)-(d). Both systems exhibit maximum $\rho_{xy}$ and $R_s$ for $x = 0.2$. In addition, $R_s(T)$ reaches maximum at $T < T_{\text{Curie}}$, suggesting the relevance of spin fluctuations. Moreover, except for $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$, $\rho_{xx}$ of the cobaltites is reduced under an external magnetic field $H$, and the magnitude of magnetoresistance, $\Delta R_H \equiv \rho_{xx}(H) - \rho_{xx}(0) / \rho_{xx}(0)$, reaches maximum near $T_{\text{Curie}}$, as shown in Figures 1(a) and 1(c). These data are consistent with the suppression of disorder spin scattering under a finite $H$. On the other hand, $\Delta R_H(T)$ changes sign near maximum $R_s(T)$ for $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$. This anomalous behavior is not yet understood.

For a given doping level $x$, the magnitude of $R_s$ in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ is much smaller than that in $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$, possibly due to stronger spin fluctuations in the latter, whose Curie temperatures are lower so that more low-spin Co$^{III}$ ions may switch to the high spin states near $T_{\text{Curie}}$. Another possibility is that carriers moving in a non-trivial spin background may acquire a “Berry phase” [4] that affects the motion of carriers in the same way as does an external magnetic field. Recent theory based on the concept of Berry phase [5] has shown that for $T < T_{\text{Curie}}$, $(R_s/R_0) \propto \exp(-E_c/(k_B T))$, where $E_c$ is the “core energy” for creating topologically non-trivial spin configurations, and that $E_c \propto T_{\text{Curie}}$. The relevance of Berry phase is consistent with our experimental observation that $R_s$ in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ is significantly smaller than that in $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$ for a given $x$, because $T_{\text{Curie}}$ and $E_c$ of Sr-doped cobaltites are larger than those of the Ca-doped cobaltites. However, the Berry phase theory for spontaneous Hall effect has been developed for $\text{La}_{1-x}\text{A}_x\text{MnO}_3$, where the Hund’s on-site exchange interaction energy ($J_H$) is much larger than the hopping energy ($t$) of the carriers. Although this assumption may be relaxed, several differences are noteworthy: $R_s$ in cobaltites is significantly larger than that in the manganites, whereas $|\Delta R_H|$ in the cobaltites is several orders of magnitude smaller; $R_0$ and $R_s$ are of the same sign in the manganites, and are opposite in the cobaltites; the conducting $t_{2g}$ electrons of the cobaltites move in a background of core electrons ($t_{2g}^2$) of opposite spin orientation, whereas the spins of conducting $e_g$ electrons in manganites are parallel to those of the core electrons ($t_{2g}^2$). These differences must be fully considered in a more complete theoretical description for the spontaneous Hall effect.

References