



# Magnetic percolation effect on the spontaneous Hall resistivity and magnetoresistance of $\text{La}_{1-x}\text{A}_x\text{CoO}_3$ ( $\text{A} = \text{Ca}, \text{Sr}; 0.1 \leq x \leq 0.5$ )<sup>☆</sup>

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## Abstract

The Hall resistivity and magnetoresistance of  $\text{La}_{1-x}\text{A}_x\text{CoO}_3$  ( $\text{A} = \text{Ca}, \text{Sr}$ ) 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. © 2000 Elsevier Science B.V. All rights reserved.

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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 on epitaxial films of ferromagnetic cobaltites  $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$  ( $0.2 \leq x \leq 0.5$ ) [3,4] reveal novel properties and a record value of  $R_s$  ( $\approx 1.4 \times 10^{-6} \text{ m}^3/\text{C}$ ) in  $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$  that cannot be explained by conventional theory. In this work, we extend our studies to epitaxial films of  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  ( $x = 0.2, 0.5$ ), and focus on the comparison of Sr-doped with Ca-doped systems, and also on the comparison of the cobaltites with the ferromagnetic  $\text{La}_{1-x}\text{A}_x\text{MnO}_3$ .

The crystal-field splitting energy in  $\text{LaCoO}_3$  is known to be larger than the Hund's energy, so that the trivalent Co-ions are primarily in the low-spin state ( $\text{Co}^{\text{III}}: t_{2g}^6 e_g^0$ ). The substitution of divalent Ca or Sr in  $\text{LaCoO}_3$  results in tetravalent  $\text{Co}^{\text{IV}}$  ( $t_{2g}^5 e_g^0$ ) ions, which stabilizes the higher-spin configurations ( $\text{Co}^{3+}: t_{2g}^5 e_g^1$  or  $t_{2g}^4 e_g^2$ ), and bulk ferromagnetism is established at  $x > 0.18$  via the double-exchange interaction [3,4]. For a given  $x$ , the fraction of higher-spin  $\text{Co}^{3+}$  increases with  $T$ . In  $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$ ,  $R_s(T \approx T_{\text{Curie}})$  reaches a record value among all known stoichiometric ferromagnetic materials [3,4]. The enhancement of  $R_s$  near the magnetic percolation threshold ( $x \sim 0.2$ ) may be attributed to the increasing spin–orbit scattering. That is,  $R_s$  is proportional to the spin–orbit coupling strength  $\lambda_{\text{so}}$ , and  $\lambda_{\text{so}} \sim [(\mathbf{k} \times \boldsymbol{\sigma}) \cdot \nabla V_c]$ , where  $V_c$  is the crystalline potential. It is known that the cobaltites consist of higher-spin hole-rich clusters embedded in a background of low-spin hole-poor matrix. Therefore, increasing  $\nabla V_c$  and spin–orbit scattering is expected near the magnetic percolation threshold, yielding enhanced  $R_s$ . This argument is consistent with the experimental data of the ferromagnetic  $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$ , as summarized in Figs. 1(a)–(b), also of the ferromagnetic  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ , as shown in Figs. 1(c)–(d). We note that both systems exhibit maximum  $\rho_{xy}$  and  $R_s$  for  $x = 0.2$ , and that  $R_s(T)$  reaches

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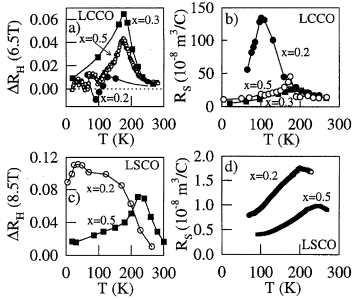


Fig. 1. Magnetoresistance and Hall resistivity of ferromagnetic  $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$  and  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ .

maximum at  $T < \sim T_{\text{Curie}}$ . Comparing  $\rho_{xx}$  with  $\rho_{xy}$ , we find that except for  $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$ ,  $\rho_{xx}$  of cobaltites decreases with increasing 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 Figs. 1(a) and (c). These data suggest the relevance of spin fluctuations and the correlation between  $\rho_{xx}$  and  $\rho_{xy}$ . However, anomalous sign change in  $\Delta R_H(T)$  is observed in  $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$  near  $T_{\text{Curie}}$  [3,4], (see Fig. 1(a)), which is not yet understood.

For a given  $x$ ,  $R_s$  of  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  is much smaller than that of  $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$ , as shown in Figs. 1(b) and (d). A plausible scenario may be related to a recent Berry phase theory for the anomalous Hall effect in ferromagnetic  $\text{La}_{1-x}\text{A}_x\text{MnO}_3$  [5], which considers a “Berry phase” [6] acquired by carriers moving in a non-trivial spin background. It is argued that the Berry phase affects the motion of carriers in the same way as does an external magnetic field, and it has been 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, with  $E_c \propto T_{\text{Curie}}$ . The relevance of Berry phase is consistent with our experimental observation in

that  $T_{\text{Curie}}$  and therefore  $E_c$  of Sr-doped cobaltites are larger than those of the Ca-doped cobaltites, so that  $R_s$  of  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  is significantly *smaller* than that in  $\text{La}_{1-x}\text{Ca}_x\text{CoO}_3$  for a given  $x$  and  $T$ . 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  $e_g$  electrons between sites of  $\text{Mn}^{3+}(t_{2g}^3 e_g^1)$  and  $\text{Mn}^{4+}(t_{2g}^3 e_g^0)$ . Although this assumption may be relaxed, particularly for the cobaltites where  $J_H$  is smaller than the crystal-field energy, 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 cobaltites, and are opposite in the manganites; the conducting  $e_g$  electrons of the cobaltites move in a background of core electrons ( $t_{2g}^5$ ) with smaller total spin ( $S = \frac{1}{2}$ ), comparing with the larger total spin ( $S = \frac{3}{2}$ ) of the core electrons ( $t_{2g}^3$ ) in the manganites. These differences must be fully considered in a more complete theoretical description for the spontaneous Hall effect.

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