

## Correlation of anomalous Hall resistivity, magnetoresistance, and magnetization in thin films of $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$

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The Hall resistivity  $\rho_H$  and magnetoresistance of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  ( $T_C \sim 360$  K) have been measured at temperatures up to 300 K in fields to 6 T. The linear correlation between the anomalous Hall coefficient  $R_s$  and the longitudinal resistivity  $\rho$  related to skew scattering is found. The change of the anomalous part is attributed to the thermal spin fluctuation that could be described by an empirical relation  $R_s \propto [M(0) - M(T)]$ . The correlation among  $R_s$ ,  $\rho$ , and  $[M(0) - M(T)]$  directly suggests that the dependence of resistivity may be described by the scattering due to thermal spin disorder. [S0163-1829(99)01841-X]

### I. INTRODUCTION

Recently, the manganese perovskites  $A_{1-x}B_x\text{MnO}_3$  ( $A$  is a rare earth,  $B$  is a divalent dopant) attained a great amount of attention, due to the colossal magnetoresistance (CMR) effect in the vicinity of the paramagnetic-to-ferromagnetic transition. The close interplay between magnetic and transport properties can be explained by the double-exchange model augmented by the Jahn-Teller effects.<sup>1</sup> According to this scenario, the nominal  $\text{Mn}^{3+}$  ion in the parent compound  $\text{LaMnO}_3$ , an antiferromagnetic insulator, has the electron configuration of  $t_{2g}^3e_g^1$ . The partial substitution of  $\text{La}^{3+}$  by the divalent ions, e.g.,  $\text{Ca}^{2+}$  in  $\text{LaMnO}_3$  provides a mixture of  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions. Among these  $3d$  electrons, the low-energy  $t_{2g}^3$  triplet contributes a local spin of  $S = \frac{3}{2}$  because of relatively poor hybridization with O  $2p$  states, while the strongly hybridized  $e_g^1$  state is either itinerant or localized depending on the local spin orientation. The  $e_g^1$  electron hopping between  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  induces the electric and ferromagnetic correlations. Therefore, the transport properties are sensitive to the interaction between the local spins and the dynamics of the  $e_g^1$  carriers. It is believed that the strong spin-dependent scattering together with its dependence on the applied field qualitatively describes the physical origin of the magnetoresistance in these materials.

Numerous measurements trying to unveil the nature of the charge transport have been published,<sup>2-9</sup> however, the scattering mechanism is still debatable. The main issue is on the correlation between the magnetic order and charge scattering. In this regard, the transverse magnetoresistance, i.e., the Hall effect, provides valuable information, e.g., the number and mobility of charge carrier, specific scattering mechanism, etc. Especially in a ferromagnetic material the embedded magnetic moments produce an anomalous Hall effect, giving the possibility of investigating the properties of a spin current.<sup>10</sup>

In the earlier studies, the origin of CMR is related to the mobility rate of the charge carrier. The carrier concentration is not influenced by the external field, and the CMR effect results from the field-induced enhancement of carrier mobility.<sup>4,6</sup> The present experimental data, however, show that the carrier concentration derived from the Hall measure-

ment is much higher than the value expected from the chemical composition.<sup>5,6,9</sup> Since the origin of the anomalous Hall effect in the manganese perovskite system is not completely known yet, the evaluation of Hall data in this means is spurious.<sup>5</sup>

The anomalous Hall effect had been discovered in some ferromagnetic metals before the advent of CMR effect in manganese perovskites. In the ferromagnetic region of these metals, a relationship

$$\rho_H(B, T) = R_0(T)B + \mu_0 R_s(T)M(B, T) \quad (1)$$

is established, where  $M(B, T)$  is the magnetization,  $R_0(T)$  and  $R_s(T)$  are the normal and anomalous Hall coefficients, respectively, and  $B$  is the applied magnetic induction. The  $R_0B$  term in Eq. (1) accounts for the ordinary Hall effect arising from the influence of the Lorentz force, while the second term  $\mu_0 R_s M$  is the contribution of the magnetic response in the materials. It is generally believed that the anomalous contribution is established by an asymmetric scattering of the localized moments on the magnetic ions. In magnetic materials, there often exists a direct correlation between the anomalous Hall coefficient and longitudinal resistivity expressed in the following form:

$$R_s(T) \propto \rho(T)^n. \quad (2)$$

Several theories explaining the intimate relations between  $R_s(T)$  and  $\rho(T)$  have been developed. Karplus and Luttinger first proposed a model to explain the anomalous Hall effect in itinerant ferromagnets.<sup>11</sup> An intrinsic spin-orbit coupling of magnetic electrons that are unequally populated in the spin-up and spin-down bands leads to an asymmetry scattering. One of the predictions of their theory is that  $R_s \propto \rho^2$  in electrically isotropic systems. Later, the theory was extended to include other scattering mechanism, e.g., scattering from impurities, lattice imperfections, and phonons. Berger further proposed that the anomalous Hall effect can result from two elementary processes:  $n=1$  for skew scattering and  $n=2$  for side jump.<sup>12</sup> This model is widely applied and discussed in the context of giant magnetoresistance (GMR) materials.<sup>13</sup> The skew scattering term, believed to rise from the spin-orbit coupling between the magnetic moment and the conduction electron, is expected to dominate in pure materials at low

temperatures. The side jump is predominant at higher temperatures and in materials with high resistivities.

A different model has been considered by Kondo: the charge carriers,  $s$  electrons, are equally distributed between states of opposite spins and the ions have a nonvanishing total spin due to the localized spin of the  $d$  electrons.<sup>14</sup> The mechanism takes into account the  $s$ - $d$  spin-spin interaction and intrinsic spin-orbit interaction of the  $d$  electrons within the magnetic ions. The skew scattering is caused by an anisotropy of the interaction between  $s$  electrons and  $d$  electrons. Kondo's expression for the anomalous Hall coefficient can be written as

$$R_s \propto \langle (M - \langle M \rangle)^3 \rangle \equiv \langle \delta M^3 \rangle. \quad (3)$$

In conventional itinerant ferromagnets, Kondo's theory is in good agreement with the results of Fe and Ni.

Later, Irkhin *et al.* considered another spin-orbit interaction, the mixed spin-orbit interaction.<sup>15</sup> Combining with all three scattering mechanisms, i.e., spin-disorder scattering, mixed and intrinsic spin-orbit interaction of the magnetic electrons, their expression for  $R_s$  can be written as  $R_s = R_i + \alpha T^3 + \beta T^4$ , which is valid for low temperatures ( $T \ll T_C$ ) and scattering by thermal spin disorder. Here  $\alpha$  and  $\beta$  are constants, which represent, respectively, the coupling strengths for the mixed spin-orbit interaction and the intrinsic spin-orbit coupling of magnetic electrons.

These theories essentially explain the anomalous Hall effect by considering a scattering mechanism for the itinerant charges together with an appropriate spin-orbit interaction, which leads to the asymmetry scattering. This effect provides another way to study the scattering mechanism from transport measurements.

Nonetheless, the observed exponent  $n$  in Eq. (2) for the manganite perovskites is uncertain. There is no direct connection between  $R_s$  and  $\rho^n$  in  $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$  compound and exponent  $n$  for  $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$  film is 1.74, as discussed by Wagner *et al.*<sup>3,4</sup> In the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  system,  $n=1$  was observed by Matl *et al.*<sup>5</sup> and  $n=2.1$  by Jakob *et al.*<sup>6</sup> Asamitsu and Tokura reported that in crystals of  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  ( $0.5 \geq x \geq 0.18$ ),  $n$  depends on the longitudinal resistivity and doping level  $x$ , approximately  $n \approx 1.2-2$ .<sup>9</sup> More experiments are needed to explain these differences.

In order to clarify these discrepancies and gain a better understanding of the CMR in the ferromagnetic state, we report a detailed Hall measurement in  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  films. A linear scaling between anomalous Hall coefficient  $R_s$  and longitudinal resistivity was found above 100 K, suggesting a skew scattering mechanism. The absence of  $R_s$  at low temperature implies that thermal spin disorder may play an important role in asymmetry scattering. The relevance of the scaling to the material transport and magnetic properties enables us to check the existing theories.

## II. EXPERIMENT

The ceramic  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  (LSMO) targets were synthesized using a conventional solid-state reaction method. Stoichiometric amounts of high purity  $\text{La}_2\text{O}_3$ ,  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ , and  $\text{MnCO}_3$  powders were mixed, ground, and calcined in an  $\text{Al}_2\text{O}_3$  crucible at 900 °C for 6 h, 950 °C for 6 h, and then 1000 °C for another 6 h in air. The resultant

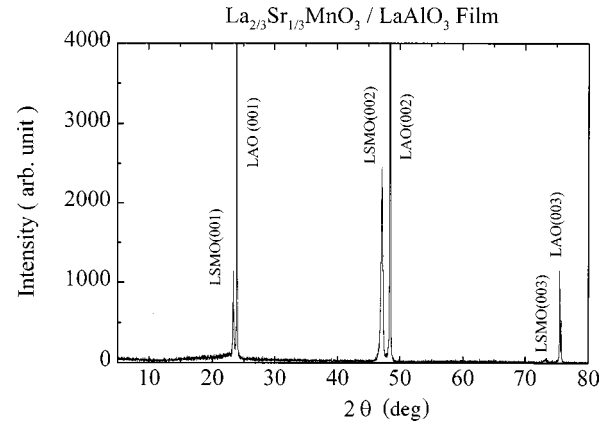


FIG. 1. X-ray diffraction patterns with Cu  $K\alpha$  radiation for  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film.

powder was then reground again and pressed into pellets, which was then sintered at 1450 °C for 6 h in flowing oxygen. The measurements were performed on epitaxial films of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  grown on (100)  $\text{LaAlO}_3$  substrates using laser ablation with a 248-nm XeF-excimer-laser pulse in 0.4 Torr oxygen atmosphere. The substrate temperature was maintained at 700 °C during the deposition. The thickness was about 200–250 nm for a deposition rate 3 nm/min. After deposition, the samples were cooled to room temperature at the rate of 10 °C/min in 1-atm oxygen atmosphere. The lattice structures of the grown films were investigated using a Rigaku Rotaflex rotating anode powder x-ray diffractometer (Cu  $K\alpha$  radiation). The dc magnetizations were measured with a Quantum Design Magnetometer MPMSR2. For measuring the Hall effect and the longitudinal resistivity  $\rho$ , the films were patterned photolithographically into a 1-mm-wide and 3-mm-long Hall bar configuration. The Hall voltage was obtained with a measuring current of 2 mA. To get rid of the extraneous contributions due to misalignment and thermoelectric effect, the Hall voltage was obtained by proper averaging of the signals measured at two reversal directions of the measuring current and the magnetic field. Since a hysteresis in low fields due to the ferromagnetic domains is observed, to obtain a uniquely defined Hall resistivity  $\rho_H(H)$  we took five sets of Hall voltages in the sweeping  $H$  ( $0 \rightarrow 6 \text{ T} \rightarrow 0 \rightarrow -6 \text{ T} \rightarrow 0 \rightarrow 6 \text{ T}$ ).  $\rho_H(H)$  is obtained by averaging the last four sets of data. Note that the hysteresis only occurred in  $H \leq 0.05 \text{ T}$ . This averaging process does not affect the data above 0.05 T.

## III. RESULTS AND DISCUSSION

A typical x-ray diffraction of the as-deposited films shown in Fig. 1 indicates a perovskite-type structure with a (100) orientation and  $c$ -axis lattice constant of 3.891 Å. The full width at half-maximum of the (100) line for all films grown was less than 0.2°, indicating a high-quality crystalline structure. X-ray-diffraction patterns showed that the films are single phase without any extra peaks due to impurities within the experimental error. The chemical composition of the films was confirmed by energy-dispersive x-ray analysis.

Figure 2 shows a representative magnetization vs temperature curve at 1 T. The field was applied perpendicular to

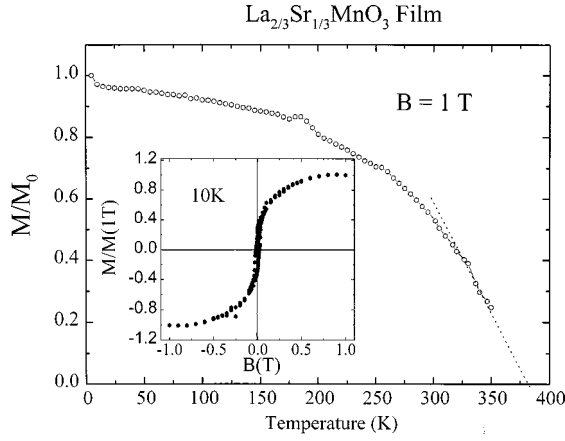


FIG. 2. Magnetization of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film at 1 T. The dashed line indicates as  $M(T)$  data linearly extrapolates to zero, it intercepts to 360 K. Inset shows the full hysteresis loop of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film at 10 K.

the film plane for the measurement. The transition between the ferromagnetic and paramagnetic states is observed corresponding to a Curie temperature  $T_C$  about 380 K determined by linearly extrapolating the  $M(T)$  data to zero.<sup>16</sup> As it is well known that the oxygen nonstoichiometry introduces hole doping, it will change the competition between the superexchange coupling and the double-exchange transfer integrals. Increasing the oxygen deficiency would strongly suppress the Curie temperature  $T_C$  while increasing the resistivity and magnetoresistance ratio.<sup>17,18</sup> The oxygen deficit of thin film is difficult to determine. However,  $T_C$  ( $=360$  K, 100 G) measured in our sample is close to those observed in high-quality crystals,<sup>9,19</sup> it suggests that the oxygen deficiency of our samples is negligible. The inset displays the full hysteresis loop at 10 K. The saturation magnetization at 4.2 K is about  $3.44\mu_B$ , close to that expected for the manganese ions. To estimate  $\mu$ , we use  $g\mu_B[f_{\text{Mn}^{4+}}S_{\text{Mn}^{4+}} + f_{\text{Mn}^{3+}}S_{\text{Mn}^{3+}}]$ , where the fraction of  $\text{Mn}^{4+}$  and  $\text{Mn}^{3+}$  are  $f_{\text{Mn}^{4+}} = \frac{1}{3}$  and  $f_{\text{Mn}^{3+}} = \frac{2}{3}$ , and the spin numbers are  $\frac{3}{2}$ , and 2 for  $\text{Mn}^{4+}$  and  $\text{Mn}^{3+}$ , respectively.  $g$  is the electron spin factor and is taken to be 2. Substituting these all, we obtain  $\mu = 3.67\mu_B$ . The diamagnetic signal of the  $\text{LaAlO}_3$  substrate have been subtracted from the sample. Unlike Asamisu and Tokura,<sup>9</sup> our as-grown films exhibit a small hysteresis loop at 10 K with a coercivity of 200 G expected from the pinning defect, demagnetization, or uniaxial anisotropy effects. At low field, magnetization rises quickly due to the magnetic domain-wall movement, and then approaches saturation at higher field probably due to the domain rotation. The saturation field is about 0.7 T at 10 K. Note in the ferromagnetic state, magnetization  $M$  for our thin films initially increases with  $H$  with a finite slope in contrast with the abrupt jump observed in single crystals whose  $M$  reaches 95% of its saturation value at 0.5 T.<sup>19</sup> We believe the difference is due to the strong pinning in films, which obstructs the spontaneous alignment of individual magnetic domains, as discussed by Matl *et al.*<sup>5</sup>

The temperature dependence of the resistivity  $\rho(T)$  of the  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film with thickness of 200 nm in zero and 4 T is shown in Fig. 3. In the ferromagnetic state, metallic conduction is observed. The magnetoresistance, de-

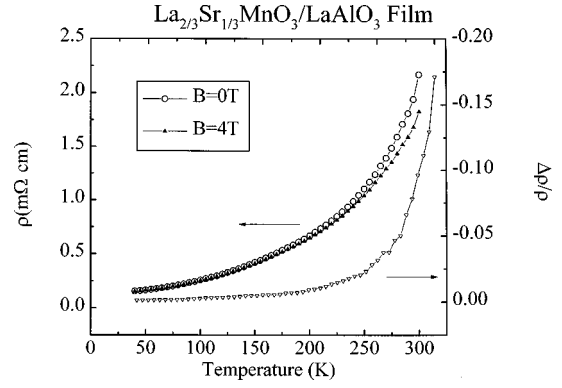


FIG. 3. The temperature dependence of resistivity for  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film at  $B=0$  and 4 T and the magnetoresistance  $\Delta\rho/\rho$ .

defined as  $\Delta\rho/\rho = [\rho(H=4\text{ T}) - \rho(0)]/\rho(0)$ , increases with temperature in our measuring range. As reported previously by Snyder *et al.*,<sup>20</sup> the resistivity can be analyzed with a fit to  $a_0 + a_2T^2 + a_{4.5}T^{4.5}$ . The temperature-independent term  $a_0$  is ascribed to scattering processes such as impurity, defect, grain-boundary, or domain-wall scattering. The  $T^2$  dependence is predicted by single-particle spin-flip excitations and the  $T^{4.5}$  term is assigned to spin-wave scattering as predicted by the double-exchange theory. Snyder *et al.* reported that the field dependence of  $a_2$  is too small to be measured and  $a_{4.5}$  decreases with increasing field. On the contrary, Mandal *et al.* observed that both  $a_2$  and  $a_{4.5}$  decrease with magnetic field.<sup>7,21</sup> The fitting data of our experiments are listed in Table I and shown in Fig. 4. The results of this analysis demonstrate that fitting parameter  $a_2$  increases but  $a_{4.5}$  decreases with increasing field, unlike Mandal *et al.* and Snyder *et al.* More fitting parameters do not significantly improve the fit. Keeping the number of free parameters to three, the fit with the coefficient of any odd powers of  $T$  becomes much poorer. The absence of linear  $T$  dependence term implies that the resistivity due to electron-phonon scattering is insignificant within this temperature range.

In Figs. 5 and 6, we present a detailed comparison of the field dependence of the longitudinal magnetoresistance  $\Delta\rho/\rho$  and Hall resistivity  $\rho_H$ . Due to the residue field and the resistivity memory effect, data below 500 G are excluded. With increasing field, these curves indicate the following trends: (a) at low temperatures,  $\Delta\rho/\rho$  varies linearly with  $H$ ; (b) the absolute magnitude of  $\Delta\rho/\rho$  increases with temperatures; (c) at high fields ( $H > 1$  T),  $\rho_H$  increases linearly with  $H$ ; (d) at temperatures higher than 100 K,  $\rho_H$  decreases steeply with  $H$  in low field, reaches a minimum at  $H \sim 0.7$  T, and then rises linearly with a slope practically independent of temperature for higher fields. Note that  $M$  is close

TABLE I. Fitting results of longitudinal resistivity  $\rho_{xx}$ .  $\rho_{xx} (\Omega \text{ cm}) = a_0 + a_2T^2 + a_{4.5}T^{4.5}$ .

Field parameter	$a_0$ ( $10^{-4} \Omega \text{ cm}$ )	$a_2$ ( $10^{-9} \Omega \text{ cm K}^{-2}$ )	$a_{4.5}$ ( $10^{-15} \Omega \text{ cm K}^{-4.5}$ )
0 T	1.779	6.956	8.676
4 T	1.564	8.885	5.725

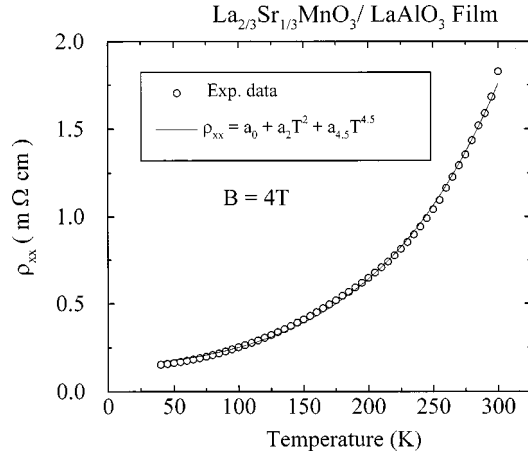


FIG. 4. The fitting results of the resistivity of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film. The solid line corresponds to  $\rho_{xx} = A + BT^2 + CT^{4.5}$  fit.

to its saturation value at  $H \sim 0.7$  T (see Fig. 2).

Unlike single crystals and bulk samples, the linearity of  $\Delta\rho/\rho$  of thin films slightly changes its slope through the region of magnetic domain rotation due to the small intergrain effect of the film's defects. It provides a clue that electron scattering at magnetic domain boundaries may not dominate transport properties, as discussed by Hwang *et al.*<sup>19</sup> On the other hand, compared with the reported behavior for the *MR* in  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ , our data are similar to single crystal. The linearity of  $\rho_H$  in low temperature ( $T < 100$  K) and high field ( $H > 1$  T) indicates that the Lorentz force dominates within these regimes. When the magnetic field exceeds 1 T, the magnetization  $M$  approaches saturation and the anomalous Hall coefficient is also nearly constant. The slope of  $\rho_H(H)$  is almost unchanged in high field. Above 1 T, the value of  $R_0$  ( $3.855 \times 10^{-10} \text{ m}^3/\text{C}$ ) corresponds to a Hall density ( $n_H = 1/eR_0$ ) of 0.91 holes per unit cell in agreement with Asamitsu *et al.*<sup>9</sup> The Hall density is much larger than 0.33 expected from the Sr substitution.

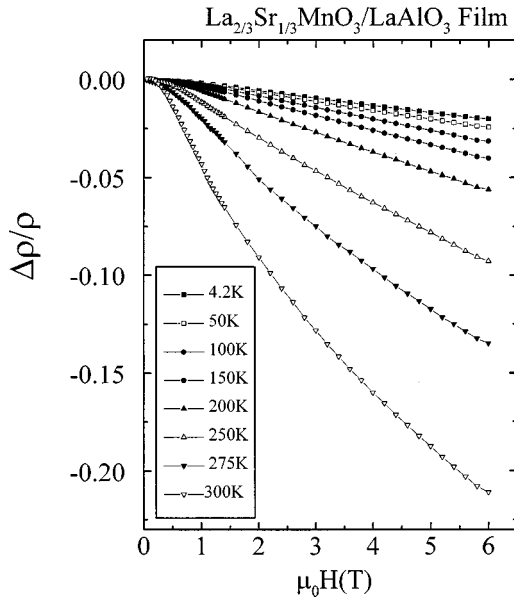


FIG. 5. The magnetoresistance  $\Delta\rho/\rho$  of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film versus field  $H$  at different temperatures.

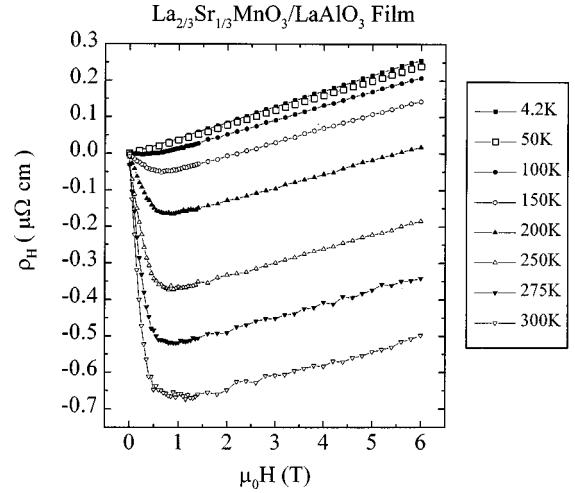


FIG. 6. The Hall resistivity  $\rho_H$  of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film versus field  $H$  at temperature from 4.2 to 300 K.

In ferromagnets,  $\rho_H$  can be fitted empirically by the Eq. (1). The applied magnetic induction is  $B = \mu_0[H + (1 - N)M]$ . Since the demagnetization factor  $N \sim 1$  in our sample geometry, the small deviation of  $B$  from  $\mu_0H$  can be ignored.

To extract  $R_s$ , we have measured  $M$  on the same sample as shown in Fig. 7. The solid dots in Fig. 7 represent the measured field dependence of the magnetization at several different temperatures and the solid lines are the results fitted with Eq. (1).  $R_s$  is the scale factor chosen to match the rela-

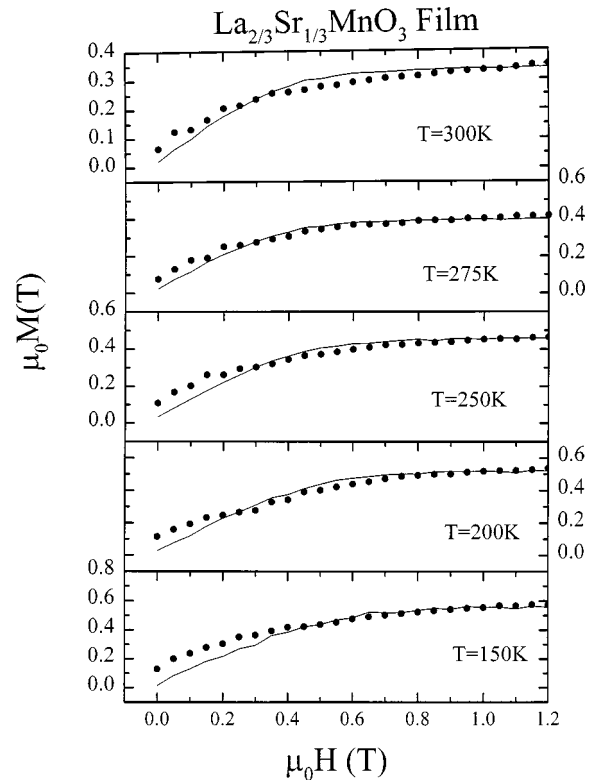


FIG. 7. The field dependence of the magnetization, solid symbols, of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film at different temperatures. The solid line is fitted by  $\mu_0 M = R_s^{-1}(\rho_H - R_0 B)$ , where the anomalous Hall coefficient  $R_s$  is the fitting factor.

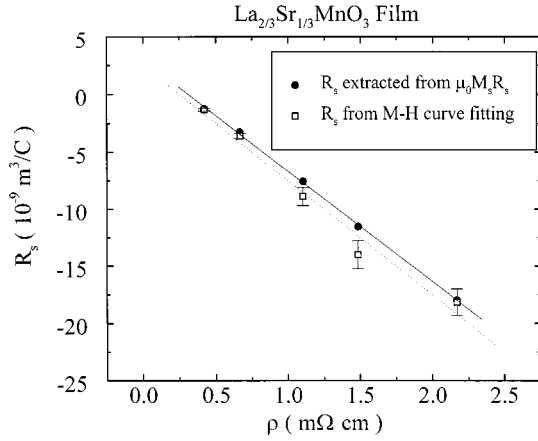


FIG. 8. The temperature dependence of  $R_s$  versus the zero-field resistivity  $\rho$  of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film. The open squares represent the  $R_s$  data extracted from the  $M$ - $H$  curve fitting and solid circles from the intercept of the  $\rho_H(H)$  curve to zero field from the linear part of high field.

tions between  $\rho_H - R_0 B$  and  $\mu_0 M$ . The error bar is estimated by the possible fitting range of  $R_s$  to  $M$ . Below 100 K,  $R_s$  is less significant. For comparison,  $R_s$  could also be obtained from the anomalous contribution  $|\mu_0 M_s R_s|$ , where  $M_s$ , the saturation magnetic moment, is taken from  $M(T)$  curve at 1 T and the spontaneous Hall effect  $\rho'_H (\equiv \mu_0 M_s R_s)$  is extracted from the intersection of the linear extrapolation of  $\rho_H(B)$  at high field. The remarkably linear relationship between  $\rho(B=0)$  and  $R_s$ , is displayed in Fig. 8. Unlike previous reports, we compare the  $R_s$  values extracted by two different ways: one from the  $M$ - $H$  fitting (solid squares) and the other from the intercept of the  $\rho_H$  curve to zero field from the linear part of high field. The deviation between these two methods probably results from the large diamagnetic contribution of the  $\text{LaAlO}_3$  substrate to  $M(T)$  at high fields. To subtract the background signal of  $\text{LaAlO}_3$ , we carefully measured a blank substrate of similar size. The  $\text{LaAlO}_3$  substrate shows diamagnetic signal, which is nearly temperature independent and increases linearly with the applied field over the whole temperature range of our interest. As the diamagnetic background of substrate becomes comparable with ferromagnetic signal of sample, the total magnetization is close to zero, and then the value calculated by the superconducting quantum interference device (SQUID) magnetometer program becomes less reliable.

The observed exponent  $n$  for the sample investigated,  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ , is nearly 1 (see Fig. 8) which is close to the results of Asamitsu *et al.*,  $n \sim 1.2$ . Similar results have also been observed by Matl *et al.* for the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  system,  $R_s(T) = \alpha \rho(0, T)$ , from 200 to 300 K in the limit  $H \rightarrow 0$ . However, Jacob *et al.* showed that  $R_s(T) \propto \rho^{2.1}$  ( $B = 9.5$  T) for their  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  films, close to the side jump value. Although the ways of Matl *et al.* and Jakob *et al.* to extract  $R_s$  are quite different, the main discrepancy between them arises from the CMR effect of the manganites' resistivity  $\rho$ . Previously existing theory to describe Eq. (2) do not take into account the magnetoresistant effect on  $\rho$  because it is a minor correction in normal metallic alloys.

Within the framework of Karplus and Luttinger, it is difficult to explain why skew scattering mechanism is quenched

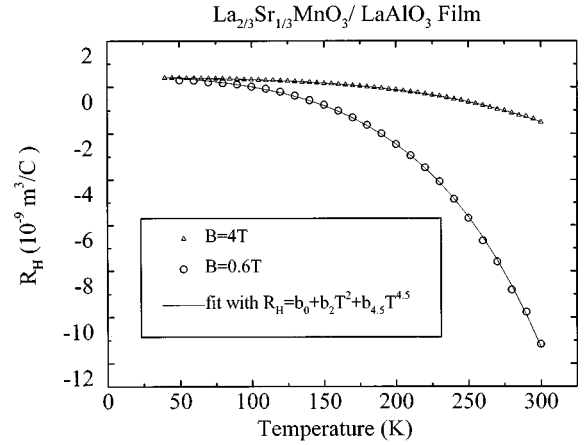


FIG. 9. Temperature dependence of Hall coefficient  $R_H$  at 4 and 0.6 T for a  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film. The solid line corresponds to the curve fitting.

below 100 K. Wagner *et al.* proposed that the total longitudinal resistivity of  $\text{Nd}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  could be written in the sense of Matthiesien's rule as  $\rho = \rho_{\text{SO}} + \rho_0$ , where  $\rho_{\text{SO}}$  represents the resistivity caused by spin-orbit-dependent scattering events and  $\rho_0$  is due to the scattering at grain boundaries, domain walls, impurities, or nonmagnetic lattice distortions. Because  $\rho_{\text{SO}}$  becomes very small below 100 K, the skew scattering is negligible,  $\rho'_H \sim 0$ . Inspecting our data (not shown here) shows that the resistivity at 4.2 K [ $\rho(4.2 \text{ K}) = 0.14 \text{ m}\Omega \text{ cm}$ ] is 44% smaller than at 100 K [ $\rho(100 \text{ K}) = 0.25 \text{ m}\Omega \text{ cm}$ ]. It suggests that in the  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  system other temperature-dependent contributions to resistivity should be included.

The disappearance of the anomalous Hall coefficient  $R_s$  at low temperatures implies that the the dominant source of the temperature dependence of  $R_s$  is due to the spin fluctuation. A strong correlation with  $R_s \propto [M(0) - M(T)]^\eta$  has been observed by Asamitsu *et al.* with  $\eta = \frac{3}{2}$ . As  $T$  approaches 0, the  $\Delta M = [M(0) - M(T)]$  vanish; the anomalous contribution disappears. To demonstrate the validity of the scaling behavior, in Fig. 9 we present the  $T$  dependence of the Hall coefficient  $R_H$  at 4 and 0.6 T.  $R_H$  shows strong temperature dependence with a sign change at around 175 K at 4 T and 100 K at 0.6 T. Higher applied field will shift the sign reversal point to higher temperature. Over the whole temperature range (4.2–300 K),  $R_H$  decreases monotonously with increasing temperature in agreement with Mandal *et al.*<sup>7</sup>

We can rewrite Eq. (1) in the following form:

$$R_H = R_0 + (\mu_0 M_s / H) R_s. \quad (4)$$

Here  $R_0$  is the normal or ordinary Hall coefficient and is nearly temperature independent. Therefore, the temperature dependence of  $R_H$  mostly results from the contribution of the anomalous part  $(\mu_0 M_s / H) R_s(T)$ . Mandal *et al.* found that the Hall coefficient fits well with  $R_H = R_0 + AT^2$  over the entire range of temperature. Since the magnetization  $[M(T) - M(0)] \propto T^2$  at low temperatures, they suggested that  $R_s$  is proportional to  $\Delta M$ . Unlike Mandal *et al.*, our data fail to fit the same formula. A  $T^{4.5}$  term is needed to correct the small deviation. The similar temperature dependence among  $R_H$ ,  $R_s$ , and  $\rho$  suggest the transport properties might

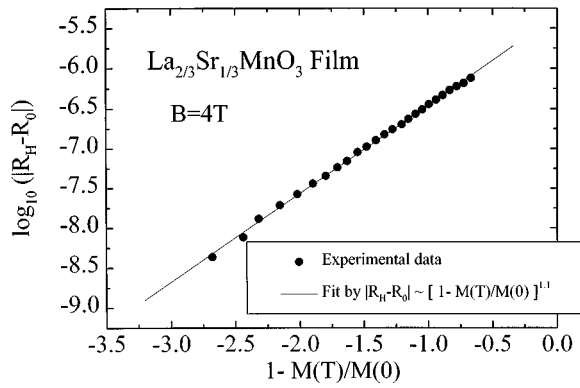


FIG. 10. The anomalous part  $(\mu_0 M_s / H)R_s$  of Hall coefficient of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film versus the change of  $[M(0) - M(T)]$  by double logarithmic scale, where  $M(0)$  denotes the saturation magnetization at zero temperature.

be governed by the same physical mechanism. It should be noted that  $R_H = b_1 + b_3 T^3$  also fit well with our data at 4 T but not the data at 0.6 T. The physical meaning behind such fitting is still not clear. In Fig. 10, the scaling factor  $\eta \approx 1$  between  $\ln(|R_H - R_0|)$  and  $\ln[1 - M(T)/M(0)]$  was observed, different from Asamitsu *et al.* and Kondo's prediction. Together with previous discussion, the correlation of  $R_s \propto \rho \propto [M(0) - M(T)]$  appears. It suggests that the longitudinal resistivity is related to the thermal spin fluctuation. The scale relation also implies a possible way to estimate the double-exchange strength under the influence of spin disorder. Applied field will significantly suppress the spin fluctuation and thus less scattering occurs, thereby producing the CMR effect.

Within the scheme of Kondo or the theory of Irkhin *et al.*,  $s$ - $d$  spin-spin interaction is necessary to give asymmetry scattering besides appropriate spin-orbit interaction. In perovskite manganite some analogy of the electronic configuration may be held, such as  $s \leftrightarrow e_g$ ,  $d \leftrightarrow t_{2g}$ . However, this analogy is superficial because the itinerant  $d$  electron  $e_g$  for the manganites has a different wave symmetry than the  $s$ -like charge carrier in early theories, which caused different spin-orbit interaction and scattering processes. Existing theories are less successful for manganite perovskite compounds due to the large magnetoresistance effect. Because there is one itinerant electron per Mn site and yet  $\text{LaMnO}_3$  is an insulator due to the strong on-site Coulomb interaction between the itinerant electrons, the manganese perovskites belong to the strongly correlated systems. There is a need for a new formalism in which the anomalous Hall effect can be calculated with the double-exchange interaction in a strongly correlated system.

In summary, we have studied the anomalous Hall effect in  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  thin film. The anomalous Hall coefficient  $R_s$  was found to linearly scale with the longitudinal resistivity, suggesting possible skew scattering. From the absence of asymmetry scattering in an ordered magnetic structure regime, we found the change of  $R_s$  could be well described as  $R_s \propto [M(0) - M(T)]$ . Although existing theory can qualitatively describe the anomalous Hall effect, it fails to quantitatively account the experimental data.

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