Giant magnetoresistance induced by spin-correlation scattering in magnetic thin films and other compounds

Liang-Jian Zou
Institute of Solid State Physics, Academia Sinica, P.O. Box 1129, Hefei 230031, China

X. G. Gong and Qing-Qi Zheng
CCAST (World Laboratory), P.O. Box 8730, Beijing, 100080 and Institute of Solid State Physics, Academia Sinica, P.O. Box 1129, Hefei 230031, China

C. Y. Pan
Physics Department, Center of Magnetism in Information Technology, Utah State University, Logan, Utah 84332

We present the study of the giant magnetoresistance effect in ferromagnetically ordered thin film and bulk based on the Hund's rule coupling between the mobile *d* electron and the core spin of Mn ions. It has been shown that the resistivity is proportional to the spin–spin correlation functions, a maximum resistivity appears near the critical point in absence of magnetic field and an applied field drives the resistivity peak to higher temperature and reduces the peak value, which is in agreement with the experiments. The giant magnetoresistance effect in thin film is attributed to the spin-correlation-dependent scattering and the low-dimensional character. © *1996 American Institute of Physics.* [S0021-8979(96)51808-2]

I. INTRODUCTION

Recently negative magnetoresistance (MR) has been found in perovskitelike ferromagnetic semiconductive thin films, such as in the films of $La_{1-x}Ba_xMnO_3$,[1] $La_{1-x}Sr_xMnO_3$,[2] $La_{1-x}Ca_xMnO_3$ (Ref. 3) and $Nd_{1-x}Sr_xMnO_3$.[4] Their MR ratios $[(R_B-R_0)/R_B]$ reported are much larger than those found in magnetic multilayers. Especially, in epitaxial $La_{1-x}Ca_xMnO_3$ and $Nd_{1-x}Sr_xMnO_3$ thin films, MR ratios are found to be $\sim 10^3$–10^4 under magnetic field $B=6$ T. More recently a giant MR ratio is also found to be in excess of 10^2 for polycrystalline La–Y–Ca–Mn–O compounds.[5] These remarkable findings can provide promising industrial applications in devices. It also brings us an interesting problem, namely, the mechanism of exhibiting such a giant MR in these ferromagnetic transition-metal–oxide compounds.

Some mechanisms[1–8] have been suggested to explain the experimental findings. These studies provide in depth investigations for the problem of interest. However, it seems to us that these proposed mechanisms do not provide a satisfactory answer. The double exchange and magnetic polaron transfer mechanism suggested by some authors[1–5] can explain qualitatively the semiconductive behavior in conduction and the negative temperature coefficient of the resistivity above T_c in the $La_{1-x}R_xMnO_3$ compounds. However, as pointed out by Millis *et al.*,[6] there exist crucial quantitative discrepancies between the experiments and the theory for the resistivity behavior in an external field, besides, it seems to be hard to form magnetic polaron in the heavy doped La–R–Mn–O and Nd–R–Mn–O systems. In the spin-disorder scattering mechanism,[7] the huge MR ratio is attributed to the alignment of the canting of manganese spins in the strong external field. However, as indicated by the experiments,[3,4] the magnetization saturates in a not large magnetic field, and increasing magnetic field further does not change the spin configurations any more, so the spin-disorder scattering mechanism is not responsible for the change of resistivity by several orders in magnitude under the high magnetic field.

In this article, we present another possible mechanism of the giant MR in these ferromagnetic semiconductive materials. The model and formalism is described in Sec. II, the theoretical curves is calculated in Sec. III, the discussion and conclusion is given in Sec. IV.

II. MODEL HAMILTONIAN

In the La–R–Mn–O or Nd–R–Mn–O compounds, the three 3*d* electrons of Mn^{+4} ions at the low level, t_g, can be considered as the localized spin S_i of the ferromagnetic background, the outer-shell 3*d* electron of Mn^{+3} ions at the high level, e_g, can hop and transfer between different Mn sites as an itinerant one, and is responsible for the electric conduction. In such a model, the outer *d* electron interacts with the core localized spin through the Hund's rule coupling, the Hamiltonian can be expressed as

$$H=H_0+V, \tag{1}$$

$$H_0=\sum_{k\sigma}(\epsilon_k-\sigma\mu_B B)c^{\dagger}_{k\sigma}c_{k\sigma}-\sum_{\langle ij\rangle}A\mathbf{S}_i\cdot\mathbf{S}_j - \sum_i g\mu_B B S^z_i, \tag{2}$$

$$V=-\frac{J}{N}\sum_{ikq}\sum_{\mu\nu}e^{i\mathbf{q}\cdot\mathbf{R}_i}\mathbf{S}_i\cdot c^{\dagger}_{k+q\mu}\sigma_{\mu\nu}c_{k\nu}. \tag{3}$$

Here ϵ_k is the energy spectrum of conduction electrons with respect to the chemical potential μ, A the effective ferromagnetic exchange constant between manganese ions, and only

 0021-8979/96/79(8)/5162/3/$10.00

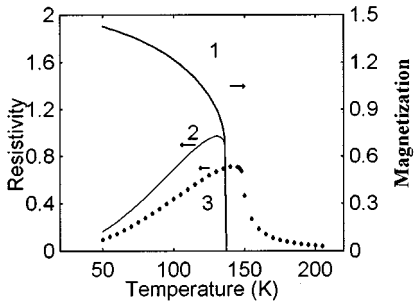

FIG. 1. Magnetization and resistivity of La–Sr–Mn–O bulks with respect to temperatures under different magnetic fields, here $J=300$ K $T_c=140$ K. (1) Magnetization in $B=2$ T, (2) resistivity in $B=6$ T, and (3) resistivity in $B=8$ T.

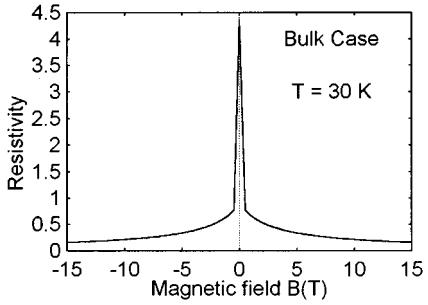

FIG. 2. Magnetoresistance of La–Sr–Mn–O bulks, theoretical parameters are chosen as $J=300$ K, $T_c=140$ K, $T=70$ K.

the nearest-neighbor interaction is considered, $g\mu_B B$ is Zeemann energy of the Mn ions in the magnetic field B; the conduction electron is scattered from state $\mathbf{k}$ spin ν to state $\mathbf{k}+\mathbf{q}$ spin μ by the localized spin $\mathbf{S_i}$ of the magnetic Mn ions. J denotes the coupling between the itinerant electron and the localized spin of Mn $3d$ electrons. One notes that under the external magnetic field and in the internal molecular field of ferromagnetically ordered state, the band of the itinerant electrons will split, this splitting will shift the position of the conduction band with respect to the chemical potential. The spectrum of the conduction electrons with state $\mathbf{k}\sigma$ is $\epsilon_{k\sigma}=\epsilon_k-\sigma(\mu_B B+J\langle S^z\rangle)$. The scattering rate of the conduction electrons scattered by the localized spins is[9]

$$\omega=\frac{2\pi}{\hbar}\sum_m |\langle f|V|m\rangle|^2\delta(E_f-E_m), \tag{4}$$

where $|f\rangle$ refers to the final equilibrium state and $|m\rangle$ the intermediate state of the system during the scattering process. Summing over all the intermediate states at a temperature of T, one gets

$$\omega=\frac{\pi}{\hbar}\frac{J^2D(0)}{4}\sum_{kq\sigma} f_{k\sigma}(1-f_{k\sigma})f_{k+q\bar{\sigma}}(1-f_{k+q\bar{\sigma}}) \times[\langle S_q^- S_{-q}^+\rangle+\langle S_q^+ S_{-q}^-\rangle+8\langle S_q^z S_{-q}^z\rangle], \tag{5}$$

where $D(0)$ is the density of states of the conduction electrons near the chemical potential, $f_k=1/[e^{\beta(\epsilon_k-\epsilon_F)}+1]$ is the Dirac–Fermi distribution function. The lifetime of the conduction electron between two scatterings is $\tau=\omega^{-1}$, and the resistivity can be obtained by the Drude formula, $\rho=m^*/ne^2\tau=m^*\omega/ne^2$, where n denotes the carrier density, m^* the effective mass of carriers. One could obtain the temperature–and field-dependent resistivity $\rho(T,B)$ as

$$\rho(T,B)=\sum_q F(q,T,B)[\langle S_q^- S_{-q}^+\rangle+\langle S_q^+ S_{-q}^-\rangle +8\langle S_q^z S_{-q}^z\rangle], \tag{6}$$

where the factor $F(q,T,B)$

$$F(q,T,B)=\frac{\pi D(0)m^*J^2}{4\hbar ne^2}\sum_{k\sigma} f_{k\sigma}(1-f_{k\sigma})f_{k+q\bar{\sigma}} \times(1-f_{k+q\bar{\sigma}}). \tag{7}$$

$F(q,T,B)$ depends on the temperature T and the magnetic field B. Accordingly, for specific doped concentration, x, the effective mass and the concentration of carriers are fixed, the resistivity is determined by the spin-correlation functions and the factor F.

III. RESULTS

In order to obtain quantitative results, the concrete forms of the transverse and the longitudinal spin–spin correlation functions over all the temperature range and in any magnetic field are needed. However, such an exact correlation function is not available for two- and three-dimensional Heisenberg ferromagnets. In this article, the approximate forms of the longitudinal and transverse correlation functions ($T<T_c$) (Ref. 8) and the longitudinal correlation function ($T>T_c$) (Ref. 9) are adopted. The resistivity and the magnetization are measured in reduced units.

A. For three-dimension cases

We first consider the bulk material of La–R–Mn–O, the lattice constant $a=3.8$ Å, and the coordinate number z is 6. The theoretical curves are evaluated for La–Sr–Mn–O compounds.

In the three-dimension case, the order–disorder transition occurs within a narrow temperature range. When approaching the critical point T_c, the spin–spin correlation function changes dramatically from the long-range correlation to short-range one. The temperature dependence of magnetization, the resistivity and the MR in the bulk La–Sr–Mn–O with the magnetic filed $B=8$ T is shown in Fig. 1, these quantities exhibit step fall when temperature (T) exceeds the Curie point T_c, which can be attributed to the weaken of the spin–spin correlation and the reduction of spin-correlated fluctuation scattering. The MR exhibits a maximum, which is similar to results of Fisher *et al.*[10]

The field-dependence of the MR below Curie temperature is shown in Fig. 2. The MR decreases monotonously with increasing magnetic field. The MR ratio in the bulk case is 1600% for $B=8$ T. The behavior of the MR above and below T_c does not agree with the experiment very well.[11] That indicates that some other scatterings are needed to be considered.

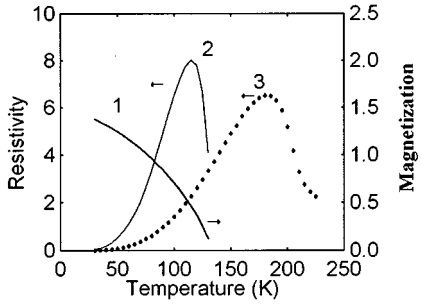

FIG. 3. Temperature dependence of magnetization and resistivity under different magnetic fields for La–Sr–Mn–O thin films where $J=500$ and $T_c=345$ K. (1) Magnetization in $B=1$ T, (2) resistivity in $B=1$ T, and (3) resistivity in $B=10$ T.

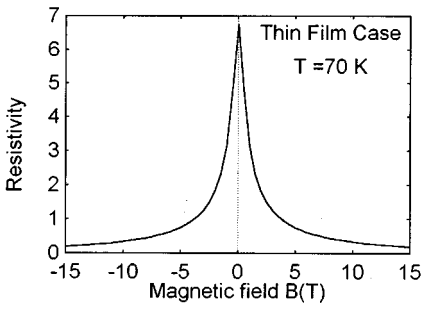

FIG. 4. Field dependence of resistivity for La–Sr–Mn–O thin films. Theoretical parameters: $T_c=240$, $J=500$, and $T=30$ K.

B. For two-dimension cases

In the quasi-two-dimensional systems, such as in thin films, the lattice constant is chosen to be the same as in the three-dimensional case, and the coordinate number is $z=4$. The temperature dependence of the magnetization and the resistivity in La–Sr–Mn–O thin film are shown in Fig. 3. The magnetization is calculated self-consistently under an external magnetic field $B=1$ T. Because of the reduction of the dimension, the transition temperature is broaded. The spin order–disorder transition occurs within a wide range (see Fig. 3). Correspondingly, the resistivities under different magnetic fields exhibit maxima within the transition region, since the spin-correlation scattering rate ω is very large near the transition point. When the magnetic field is increased, the resistivity peak moves to higher temperature and the maximum value is reduced (see curves 2 and 3 in Fig. 3). This could be interpreted as that a strong field aligns all spins in the systems and suppresses the correlation between spin at different sites, thus reduces the spin-correlated fluctuation scattering between the conduction electrons and the localized spins. These theoretical results agree with the experiments very well.[1–4]

The field-dependence of the MR for a quasi-two-dimension system is shown in Fig. 4. One can see that the magnetoresistance also decreases monotonously with increasing field in the strong field region. This fact coincides with the experiments.[1–4] The MR ratio $\Delta R_B/R_B$ in field $B=6$ T can be as high as 1000%.

IV. DISCUSSION AND CONCLUSION

The resistivity due to the interaction Eq. (3) was studied for bulks by Fisher *et al.*[11] Fisher's work doesn't consider the spin-splitting of the conduction electrons caused by the external magnetic field and the internal molecular field, which may be important for the MR effect in an external magnetic field and magnetically ordered state. Our study has shown that the spin correlation scattering is essential in the thin films and should be responsible for the giant MR near T_c where the long-range or the short-range spin–spin correlations and the spin fluctuation are large.

From this study we can summarize several results. (a) If the measurement temperature is below or above far away from the critical point T_c, the MR change ΔR_B is small. However, the MR behaviors below and above T_c may be dramatically different since the former is dominated by the fluctuation scattering of the long-range spin-correlation, the latter is by that of short-range correlation. (b) If the critical transition range is wide, a giant MR could be observed within a wide temperature range, this property is useful for the applications. It suggests us that if we want to find a new kind of material which could be used in room temperature for magnetic recording and sensitive detect, then the Curie critical point of this material should be near the room temperature, and it should be in a low-dimension system, such as in thin film. It also indicates that in the case of magnetic multilayers, if measured temperatures near T_c, a more high giant MR may be found. Further experiments are extremely desired to verify these predictions.

ACKNOWLEDGMENTS

This work is financially supported by the Grants of the NNSF of China and the LWTZ of CAS. One of the authors (Q.-Q. Z.) is grateful for support from the CMIT at Utah State University.

[1] R. M. Kusters, J. Singleton, D. A. Keen, R. McGreevy, and W. Hayes, Physica B **155**, 362 (1989).
[2] R. Von Helmolt, J. Wecker, B. Holzapfeil, L. Schultz, and K. Samwer, Phys. Rev. Lett. **71**, 2331 (1993); R. Von Helmolt, J. Wecker, K. Samwer, L. Hampt, and K. Barner, J. Appl. Phys. **76**, 6925 (1994).
[3] S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994); J. Appl. Phys. **76**, 6929 (1994); M. McCormack, S. Jin, T. H. Tiefel, R. M. Fleming, J. M. Phillips, and R. Ramesh, Appl. Phys. Lett. **64**, 3045 (1994).
[4] G. C. Xiong, Q. Li, H. L. Ju, S. N. Mao, L. Senpati, X. X. Xi, R. L. Greene, and T. Venkatesan, Appl. Phys. Lett. **66**, 1427 (1995).
[5] S. Jin, H. M. O'Bryan, T. H. Tiefel, M. McCormack, and W. W. Rhodos, Appl. Phys. Lett. **66**, 382 (1995).
[6] A. J. Millis, P. B. Littlewood, and B. I. Shraiman, Phys. Rev. Lett. **74**, 5144 (1995).
[7] N. Furukawa, J. Phys. Soc. Jpn. **63**, 3214 (1994).
[8] R. M. White, *Quantum Theory of Magnetism*, Springer Series (McGraw–Hill, New York, 1970).
[9] R. A. Tahir-Kheli, *Phase Transition and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1975), Vol. 5B.
[10] M. E. Fisher and J. S. Langer, Phys. Rev. Lett. **20**, 665 (1968).
[11] A. Urushibara, Y. Moritomo, T. Arima, A. Asamitsu, G. Kido, and Y. Tokura, Phys. Rev. B **51**, 14 103 (1995).