Electrical transport in manganite granular systems

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A systematic investigation of magnetic and transport properties is carried out for $La_{2/3}Ca_{1/3}MnO_3$ granular system. Based on the magnetization measurement, it is argued that oscillatory indirect magnetic exchange coupling via the boundary material with thickness *s* sandwiched between the neighboring grains exists in polycrystalline manganites. By introducing a distribution function of *s* and averaging the conductivity deduced from the spin-polarized tunneling model for the neighboring grains, we obtain a general expression for the temperature dependence of resistance that can reproduce the experimental data observed in polycrystalline manganites. The present model provides information on the microscopic transport mechanism.

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The perovskite manganites of type $La_{2/3}Ca_{1/3}MnO_3$ have stimulated considerable scientific and technological interest because of their exotic magnetic and electronic properties.[1–3] Most notable of these properties are the paramagnetic-to-ferromagnetic (PM-FM) transition at Curie temperature T_C, the insulator-to-metal (I-M) transition at the temperature T_{IM}, and the so-called colossal magnetoresistance (CMR)—a substantial reduction in resistivity upon application of magnetic fields. For single crystalline manganites, T_{IM} is shown to be almost the same as T_C; one, therefore, believes that the transition to metallic behavior is due to the FM ordering of Mn spins as indicated in the double-exchange model.[4] Growing attention is being paid to polycrystalline manganites in which the grain boundary effects dramatically modify their physical properties.[5] T_{IM} of polycrystalline samples is usually lower than T_C and the difference between T_C and T_{IM} increases with decreasing grain size.[6,7] Polycrystalline manganites also exhibit CMR effect, however, another kind of MR (i.e., the intergrain MR) is superimposed on the CMR effect.[7]

Despite the fact that the CMR has been intensively studied theoretically, the physical mechanism is still controversial. Experimentally, the CMR is found to be a peaked function of T[2,7,8] and the maximum value appears near T_{IM} but not T_C. In all samples showing CMR, the CMR near T_{IM} is found to exhibit *T*-dependent behavior similar to that for the resistance (R)-versus-T dependence measured in zero magnetic field, suggesting the same underlying physical origin of both the CMR and the transport process.[8] It is, therefore, needed to study the transport mechanism in order to understand the physical origin of the CMR.

The transport mechanism is not very well understood. Especially, the nature and the role of the grain boundary insulating barrier of structural origin on the transport behavior is not clear. In this paper we attempted to seek an answer to this question in the particular context of $La_{2/3}Ca_{1/3}MnO_3$ granular system. Similar to that observed in FM multilayer films,[9,12] we propose that the magnetic exchange coupling via the boundary material sandwiched between the neighboring grains shows an oscillatory behavior with boundary thickness *s*. Based on this assumption, an explanation is presented for the observation of zero magnetization without magnetic field in polycrystalline manganites. By averaging over the distribution of *s* to the conductivity deduced from the spin-polarized tunneling model, we propose a model that can reproduce the temperature dependence of resistance observed in polycrystalline manganites.

The polycrystalline $La_{2/3}Ca_{1/3}MnO_3$ sample was prepared by the sol-gel method[6] followed by a sintering treatment at 1100°C for 12 h. The x-ray diffraction analysis confirmed that the sample is single phase of perovskite crystalline structure. In Fig. 1 we present the pattern of surface distribution over a 4×4 μm^2 area revealed by the atomic force microscopic analysis. It can be found that the sample is a typical granular one with an average grain diameter $d\sim300$ nm. Electronic transport and magnetic properties were measured in a commercial Physical Property Measurement System (Quantum Design PPMS).

Shown in Fig. 2, by open circles, is the R vs T dependence

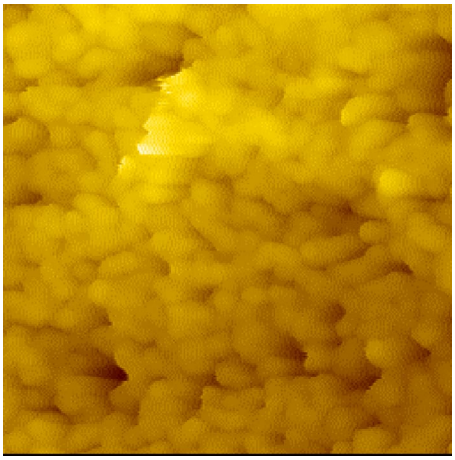

FIG. 1. (Color online) Grain distribution over a 4×4 μm^2 area revealed by the atomic force microscopic analysis for polycrystalline $La_{2/3}Ca_{1/3}MnO_3$.

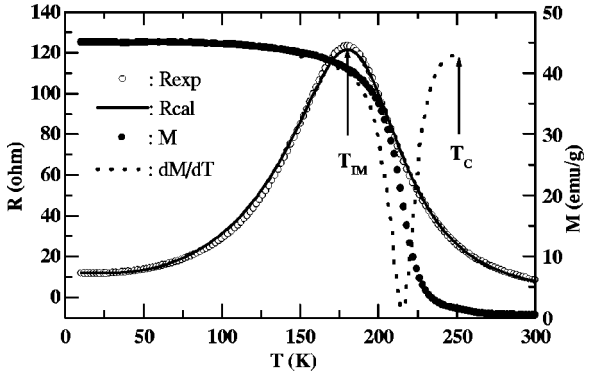

FIG. 2. Temperature dependence of resistance (open circles) in zero field and magnetization (solid circles) in a 0.1 T field, measured in polycrystalline $La_{2/3}Ca_{1/3}MnO_3$. dM/dT is indicated by the dot line. The solid line is the fit detailed in the text.

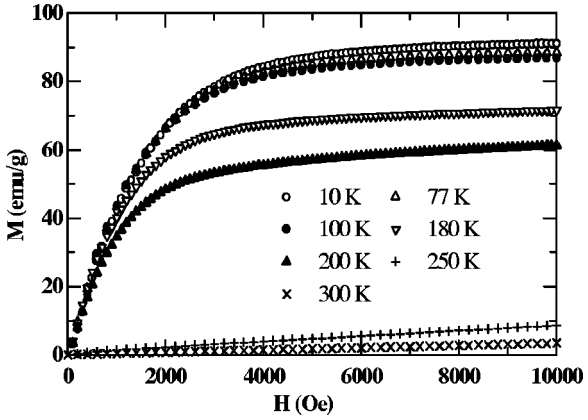

FIG. 3. Field dependence of magnetization for several selected temperatures measured in polycrystalline $La_{2/3}Ca_{1/3}MnO_3$.

measured in zero field. T_{IM} is determined to be ~ 180 K from the resistance peak. On cooling from $T > T_{IM}$ to $T < T_{IM}$, the temperature dependence of R changes from insulating behavior of $dR/dT < 0$ to metallic one of $dR/dT > 0$. Magnetization M as a function of T is measured in a field of 0.1 T, which is also displayed in Fig. 2 by solid circles. T_C is determined to be ~ 250 K below which the change in dM/dT clearly occurs, as indicated by dot line in Fig. 2. The T_C-value thus obtained is very close to the Curie temperature commonly reported on $La_{2/3}Ca_{1/3}MnO_3$ single crystals. Previous studies[7] revealed that T_C remains constant for all the samples with different grain sizes. It implies that T_C is intrinsic to perovskite manganites, and the grain size has no effect on T_C.

An essential fact should be noted in the measurement of M vs T dependence. For the 0.1 T field, the maximum magnetization appearing at low temperatures is only ~ 45 emu/g, which is about half of the saturation value ~ 93 emu/g estimated by assuming a complete FM alignment of all Mn spins. The maximum value in the measured magnetization is further decreased with decreasing applied field. This points out that the magnetization tends to vanish with decreasing the field to zero. As is well known, a ferromagnet has a spontaneous magnetization $M(T)$ even in zero applied magnetic field. This, however, cannot be obtained directly in experiments since the measurement for $M(T)$ is always performed when a field is applied. In order to obtain $M(T)$ in zero field, we have performed the measurement of magnetization as a function of applied field at various constant temperatures. In Fig. 3 we display the field dependence of magnetization at several constant temperatures. The results indicate basic features similar to that observed in granular ferromagnets: The high-T regime, as seen at $T = 250$ or 300 K, shows typical PM behavior; while for the low-T range, M first shows a sharp increase and then tends to saturate with increasing H. In all the cases, M is found to be zero when no field is applied. Similar results are commonly reported on polycrystalline (even single crystalline) manganites.[13] This queries the validity of any explanation of the metallic conductivity due to the long-range FM ordering below T_C.

As seen in Fig. 1, the polycrystalline manganite is a typical granular system. A thin boundary layer with thickness s should be present around each grain. The physical property of the grain boundary region should be different from that inside the grains, probably due to (i) the deviation of stoichiometric composition, (ii) the termination of crystalline structure, and (iii) the change of Mn environment at the grain boundaries. Actually, the following viewpoint has been widely accepted, namely, some insulating and nonmagnetic material present should be at the grain boundary region. Therefore, it is reasonable to consider that the grain (the ith grain, say) below T_C is a ferromagnet with a spontaneous magnetic moment $\mathbf{M}_i$ due to the FM alignment of Mn spins with the help of the double-exchange interaction. Nevertheless, it is likely that without external magnetic field, these magnetic moments are arbitrarily oriented, giving no effective contribution to the total magnetization, namely, $\mathbf{M}(T, H=0) = \Sigma_i \mathbf{M}_i = 0$.

The thermal energy can cause the random alignment of magnetic moments. This consideration, however, cannot explain the experimental observation of zero magnetization. In truth, even at temperatures down to the lowest available temperature (~ 2 K in our laboratory), where the low thermal energy is insufficient to affect the alignment of magnetic moments, the measured magnetization is still zero in the case of zero field. We therefore need to consider the other effect. Here a plausibility argument for the observation of $\mathbf{M}(T, H=0) = 0$ is presented by taking into account the exchange-coupling constant J between the neighboring FM grains as a function of s. For a real polycrystalline sample, both d and s would follow a wide distribution. For the sake of simplicity, we assume that the grains are roughly uniform in size so that each grain has the same magnetic moment $\mathbf{M}_i$ (the corresponding effective spin is represented by $\mathbf{S}_i$), but s can vary from one pair of grains to the next. We then expect that the exchange coupling energy of grains i and j is given by

$$E_{ij} = -2J(s)\mathbf{S}_i \cdot \mathbf{S}_j . \tag{1}$$

The earlier studies[9–12] on two FM layers separated by the nonmagnetic transition metal generally showed an oscillatory behavior in J with s. For instance, J in Co/Cu multilayer structures varies from $J > 0$ (FM coupling) to $J < 0$ (antifer-

romagnetic coupling) on increasing s from ~ 1 nm to ~ 2 nm. Further studies revealed that such an oscillatory in J is a general phenomenon for the exchange coupling via transition metals sandwiched between FM layers.[12] We, therefore, expect that such an oscillatory in J also exists in polycrystalline manganites. A random distribution of grain pairs with different (FM and antiferromagnetic) couplings gives zero resultant magnetic moment, therefore $\mathbf{M}(T,H=0)=0$.

We now turn to the discussion of electrical conductivity in polycrystalline manganites. For a granular FM metallic system, the electrical conductivity is usually understood in terms of the spin-polarized tunnelling model proposed firstly by Helman and Abeles.[14,15] This model gives the temperature dependence of the conductivity as

$$\sigma \propto e^{-2s\chi}\left[\frac{1}{2}(1+P)e^{-(E_c+W/2k_BT)} + \frac{1}{2}(1-P)e^{-(E_c-W/2k_BT)}\right], \tag{2}$$

where $\chi=\sqrt{2\mu U/\hbar^2}$ (U the barrier height, μ the effective mass of charge carrier) and P is the spin-polarization of the charge carriers. E_c is the charging energy which can be approximately expressed[16] in terms of d and s as $E_c \approx (e^2/4\pi\epsilon_0 d)[(s/d)/(1/2+s/d)]$. W is the magnetic-dependent energy barrier which is introduced when the magnetic moments of the neighboring grains are not parallel.

As discussed above, polycrystalline manganite samples at $T<T_C$ do actually behave as granular FM metallic systems. The manganite samples, on the other hand, show a nearly complete spin-polarization of the charge carriers[17] because a relatively narrow majority-carrier conduction band is completely separated from the minority band by a large Hund's energy and an exchange energy, implying $P\approx 100\%$. For the polycrystalline sample used in the present study, $E_c/2k_B$ is estimated to be much smaller than ~ 1 K from the dependence $E_c \approx (e^2/4\pi\epsilon_0 d)[(s/d)/(1/2+s/d)]$ due to $s \ll d \sim 300$ nm. For the temperature range studied, we, therefore, omit the E_C contribution to the conductivity. When the spin-polarized tunneling model is applied to two manganite grains i and j separated by insulating boundary material, the conductivity can be approximated as

$$\sigma \propto e^{-2s\chi-(W/2k_BT)}. \tag{3}$$

At $T>T_C$, the conductivity has the general form of thermal activation, i.e., $\sigma \propto e^{-E_g/k_BT}$, where E_g is the activation energy. On cooling (warming) from T_C^+ (T_C^-), the conductivity should be continuous, implying $2E_g=W(T=T_C)$. We, therefore, can extend Eq. (3) to the PM phase if letting $E_g = W(T=T_C)/2$. Equation (3) is obtained only for the two neighboring grains. In order to obtain the total conductivity of the sample, the expression shown in Eq. (3) has to be averaged over the distribution of s. The total conductivity can be then written as

$$\sigma \propto \int_0^\infty D(s)e^{-2s\chi-(W/2k_BT)}ds, \tag{4}$$

where $D(s)$ is a distribution function of s and it has the general feature of vanishing in the limits of $s\to 0$ and $s\to\infty$, and $\int_0^\infty D(s)ds=1$.

Based on the above argument of magnetic coupling properties of the neighboring grains, it is reasonable to assume that the most probable value of s is located at $s\sim s_1$ and $s\sim s_2$, respectively, where $s_1<s_2$. If $D_i(s)$ represents the distribution function near s_i ($i=1,2$), the total distribution function can be then viewed as the sum of both functions, i.e., $D(s)=D_1(s)+D_2(s)$. Equation (4) then becomes

$$\sigma \propto \int_0^\infty D_1(s)e^{-2s\chi-(W/2k_BT)}ds + \int_0^\infty D_2(s)e^{-2s\chi-(W/2k_BT)}ds. \tag{5}$$

Clearly, $D_i(s)$ is a peaked function and the maximum value of $D_i(s)$ occurs at $s=s_i$. When s deviates from s_i, $D_i(s)$ quickly decreases to zero. Therefore, $W(s)$ appearing in the first and second terms of Eq. (5) can be, approximately, replaced with $W(s_1)$ and $W(s_2)$, respectively. For $s\sim s_1$, the magnetic moments of the neighboring grains below T_C are ferromagnetically coupled, leading to a reduction in the magnetic-dependent energy barrier. In this case, this barrier is often expressed as $W(s_1)=W_1(1-m^2)$, where W_1 is a T- and H- independent constant, $m=\sqrt{\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle/S^2}$ is the magnetization normalized to the saturation value for each grain under consideration and $\langle\ldots\rangle$ indicates statistical averages. For $s\sim s_2$, the magnetic-dependent energy barrier is approximated as a constant, i.e., $W(s_2)=W_2$, due to the antiferromagnetic coupling of magnetic moments of the neighboring grains at $T<T_C$. Based on these discussions, the total conductivity can be rewritten as

$$\sigma \propto \left(\int_0^\infty D_1(s)e^{-2s\chi}ds\right)\times e^{-W_1(1-m^2)/2k_BT} + \left(\int_0^\infty D_2(s)e^{-2s\chi}ds\right)\times e^{-W_2/2k_BT}. \tag{6}$$

Letting $\rho_1=e^{W_1(1-m^2)/2k_BT}$, $\rho_2=e^{W_2/2k_BT}$, the total resistance is finally given by

$$1/R=A_1/\rho_1+A_2/\rho_2, \tag{7}$$

where A_i is a constant proportional to $\int_0^\infty D_i(s)e^{-2s\chi}ds$ ($i=1,2$).

Equation (7) is similar, in form, to that obtained by considering two parallel conduction channels[18] with low (metallic below T_C) resistivity ρ_1 and high (always insulating) resistivity ρ_2, respectively. In the present situation, however, it is derived from the more general consideration. It must be emphasized that the present model is derived from the spin-polarized tunneling and the thermal activation. Therefore, the present model provides information on the microscopic transport mechanism. To be convincing, we must show that based on this model we can explain, quantitatively, the experimental observations for the temperature dependence of resistance measured in polycrystalline manganites. Continu-

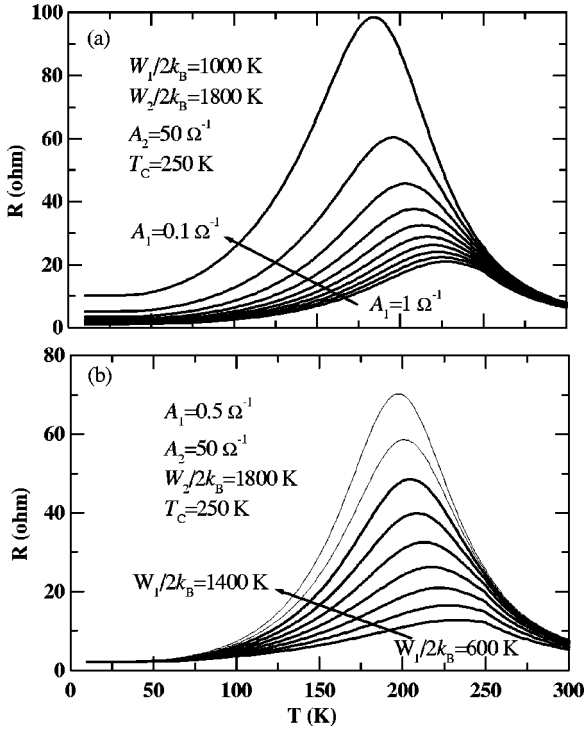

FIG. 4. R vs T curves calculated for polycrystalline $La_{2/3}Ca_{1/3}MnO_3$ with $T_C \sim 250$ K by decreasing A_1 from 1 to 0.1 in steps of 0.1 (a) and by increasing $W_1/2k_B$ from 600 K to 1400 K in steps of 100 (b).

ous line in Fig. 2 is the fit to the data measured in the sample under investigation by using $A_1 = 0.085\Omega^{-1}$, $A_2 = 50\Omega^{-1}$, $W_1/2k_B = 1050$ K, $W_2/2k_B = 1800$ K and $m(T)$ calculated in terms of the mean-field self-consistency equation with $J = 1.84$ and $T_C = 250$ K. It can be found that a satisfactory fit to the data is indeed obtained for the whole temperature range, covering from high-T insulating region to low-T metallic one.

Clearly, the conductivity behavior is strongly affected by the grain boundary effect. There are various factors that can increase the grain boundary effect. In the present approach, this effect is phenomenologically considered through both parameters A_1 and W_1. Increasing W_1 increases the height of the magnetic-dependent energy barrier, causing a decrease of the FM coupling between neighboring grain moments. On the other hand, $A_1 \propto \int_0^\infty D_1(s) e^{-2s\chi} ds$ is determined by distribution function $D_1(s)$ and the tunneling matrix over this distribution. Since the most probable value of s is located at $s \sim s_1$ for distribution $D_1(s)$, $e^{-2s\chi}$ in expression A_1 can be approximately replaced with $e^{-2s_1\chi}$. One then, approximately, has $A_1 \propto \int_0^\infty D_1(s) ds$ which is proportional to the volume fraction of the metallic region. Decreasing A_1 is therefore equivalent to a decrease in volume fraction of the metallic region. These discussions are summarized by considering two sets of R vs. T curves calculated in terms of Eq. (7) for polycrystalline $La_{2/3}Ca_{1/3}MnO_3$ with $T_C \sim 250$ K. The first set of R vs T curves, as indicated in Fig. 4(a), is calculated by decreasing A_1 from 1 Ω^{-1} to $0.1\Omega^{-1}$ while fixing parameters A_2, $W_1/2k_B$, and $W_2/2k_B$ at 50 Ω^{-1}, 1000 K, and 1800 K, respectively. The other set of R vs T curves, as displayed in Fig. 4(b) is obtained by increasing $W_1/2k_B$ from 600 to 1400 K but fixing parameters A_1, A_2, and $W_2/2k_B$ at $0.2\Omega^{-1}$, $50\Omega^{-1}$ and 1800 K, respectively. It can be found that although T_C is fixed at 250 K, the I-M transition is gradually moved toward the low-T region with increasing grain boundary effect. Associated with the decrease of T_{IM}, the peak resistance is largely increased, and the low-T resistance is also increased. All these features are similar to that commonly observed in polycrystalline $La_{2/3}Ca_{1/3}MnO_3$.

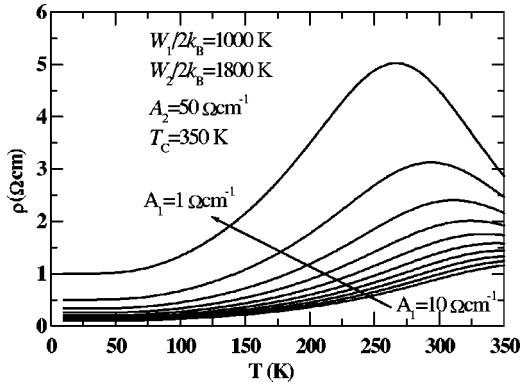

FIG. 5. Temperature dependence of resistivity calculated for polycrystalline $La_{2/3}Sr_{1/3}MnO_3$ with $T_C \sim 350$ K by decreasing A_1 from 10 to 1 in steps of 1 but fixing A_2, $W_1/2k_B$, and $W_2/2k_B$ at 50, 1100 K, and 1800 K, respectively.

The present model can also be applied to yield experimental features of electronic transport observed in the other manganite perovskites. Here we take polycrystalline $La_{2/3}Sr_{1/3}MnO_3$ with $T_C \sim 350$ K as an example. In order to obtain resistivity $\rho(T)$ in unit of Ωcm, both A_1 and A_2 are in units of Ωcm^{-1}. Indicated in Fig. 5 is a set of ρ vs T curves calculated by the present model for polycrystalline $La_{2/3}Sr_{1/3}MnO_3$ with $T_C \sim 350$ K, where A_1 changes from 10 to 1 while parameters A_2, $W_1/2k_B$, and $W_2/2k_B$ are fixed at $50\Omega\text{cm}^{-1}$, 1000 K, and 1800 K, respectively. The results show that for samples with higher A_1, which is equivalent to a larger volume fraction of the metallic region, the metallic conductivity is observed over the whole temperature range below T_C. With decreasing volume fraction of the metallic region the I-M transition is observed near room temperature. On further decreasing the volume fraction of the metallic region, the I-M transition is gradually moved toward the low-T region. Associated with the decrease of T_{IM}, the resistivity peak is clearly increased, and the low-T resistivity is also increased. All these features are similar to that commonly observed in polycrystalline $La_{2/3}Sr_{1/3}MnO_3$.

Letting $s_i = 0$ and $W_i = W$ $(i = 1,2)$ in Eq. (6), the present model can be extended to the case of single crystals. At $T > T_C$, $m = 0$, Eq. (6) is reduced to the case of $\sigma \propto e^{-W/2k_BT}$, indicating a thermal activation insulating behavior. At $T < T_C$, the sample is composed of small regions called domains or polarons. The directions of magnetization of different domains (polarons) need not be parallel. If the magnetic moments of the neighboring domains (polarons) are parallel, the conductivity is proportional to $e^{-W(1-m^2)/2k_BT}$. Otherwise, the conductivity is always insulating, i.e., $\sigma \propto e^{-W/2k_BT}$. Representing the number densities

of domain (polaron) pairs with parallel and antiparallel moment alignments as p and (1-p), respectively, the total conductivity for single crystalline manganites can be expressed as $\sigma \propto pe^{-W(1-m^2)/2k_BT} + (1-p)e^{-W/2k_BT}$.

Finally, based on Eqs. (6) and (7), we present a possible discussion on the magnetoresistance effect observed in polycrystalline manganites. The application of magnetic fields decreases ρ_1 near T_C due to the field-induced increase in m. ρ_2 near T_C is high in zero field due to the antiferromagnetic coupling of magnetic moments of the neighboring grains. When a sufficiently high field is applied, the field aligns magnetic moments of the neighboring grains, leading to a large reduction in ρ_2. Both contributions cause a substantial reduction in R and hence the CMR. At $T \ll T_C$, ρ_1 is basically independent of the field due to the nearly complete FM alignment of magnetic moments of the neighboring grains distributed over $D(s_1)$. But the application of magnetic fields can cause a reduction in ρ_2 due to the field-induced rotation of magnetic moments from grains distributed over the $D(s_2)$. Therefore, even at $T \ll T_C$, polycrystalline manganites would also show a low-T magnetoresistance behavior. These predictions are in qualitative agreement with experimental data obtained in polycrystalline manganites.

In summary, we have proposed a model for describing the electronic transport behavior in polycrystalline manganites. The basic physical idea is based on the assumption of oscillatory indirect magnetic exchange coupling via the boundary material with thickness s sandwiched between the neighboring grains. By introducing a distribution function of s and averaging the conductivity deduced from the spin-polarized tunneling model for the neighboring grains, we obtain a general expression for the temperature dependence of resistance that can reproduce the experimental data observed in polycrystalline manganites. The magnetoresistance behavior (including the CMR near T_C and the low-T magnetoresistance) observed in polycrystalline manganites can be also well explained by the present model.

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