Theory of intergranular magnetoresistance in nanometric manganites

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We present a self-consistent effective medium theory for the magnetotransport in nanometric manganites on the basis of the bond-disordered resistor network. The transport process here is assumed to be controlled by both the charging energies of grains and the spin-polarized tunneling through grain boundaries. The effects of network and band-bending at the grain boundaries on the magnetotransport are investigated. Our results are compared with the experiments on nanometric $La_{2/3}Sr_{1/3}MnO_3$ and $La_{2/3}Ca_{1/3}MnO_3$; good agreements are found.

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I. INTRODUCTION

Intergranular magnetoresistance (IMR) in half-metallic granular systems has attracted great interest since the discovery of low-field magnetoresistance in polycrystalline manganites.[1–5] IMR is characterized by a negative variation of resistance upon the application of an external magnetic field. Two regimes are usually observed. At a low field, typically some hundreds of oersted, a rapid decrease of resistance is observed, which can amount to 56% in dilute CrO_2 powder compacts[3] at low temperatures. At higher fields, a smoother, almost linear variation is found. These two regimes cross over at the field for which near magnetic saturation is achieved. Among these magnetic oxides, e.g., mixed-valence manganites,[2] chromium dioxides,[3] double perovskite manganites[4] and magnetite,[5] mixed-valence manganites of the type $A_{1-x}B_xMnO_3$ (A=La, Nd; B=Ca, Ba, Sr) are undoubtedly one of the most studied for their remarkable transport properties. They are generally believed to be double-exchange ferromagnets and, therefore, have a nearly 100% spin-polarization (P) below the Curie temperature T_c. By comparing the magnetotransport in single-crystalline $La_{0.7}Sr_{0.3}MnO_3$ and polycrystalline ceramics in detail, Hwang *et al.*[2] were the first to propose a model to explain the IMR. They attributed IMR to spin-polarized tunneling through the insulating grain boundaries in polycrystalline ceramics. And the high degree of spin-polarization of these materials enhances the tunneling magnetoresistance effect. Despite the apparent applicability of the tunneling model, there are a number of features that cannot be readily understood within this picture. Among them, the large magnitude of high field magnetoresistance (HFMR), which sometimes exceeds the low-field response, is more difficult to understand.[6]

The properties of surface and interface in manganites are thought to be crucial in determining the magnetotransport in these materials. To explore the LFMR limit in polycrystalline manganites, Balcells *et al.* have systematically studied the grain size dependence of magnetotransport and pushed the grain size down to the nanometer scale.[7] They have found that LFMR is almost saturated in the nanometer scale while the HFMR progressively rises with the decrease of grain size. In the sample with the smallest grain size, the upturn of the low-temperature resistivity is observed and understood as a result of the existence of an intergranular Coulomb gap which adds to the tunneling barrier. On the other hand, Andres *et al.* have compared the effects of grain size and connectivity on the magnetotransport in granular $La_{0.7}Ca_{0.3}MnO_3$.[8] A two-channel model (metallic and insulating) is proposed to explain both the LFMR and HFMR. The decrease of resistance in the high fields is modeled by the opening of new metallic conduction channels in parallel which is due to the ordering of Mn spins blocked at the grain surface. They concluded that it is the connectivity rather than the grain size that determines the whole transport properties in polycrystalline manganite thin film. The same conclusion is drawn by Garcia-Hernandez *et al.* in the experiment on the low-temperature resistance of granular films of manganites with nanometric grain sizes.[9] To explain the upturn in the low-temperature resistance and magnetic field dependent of the Coulomb gap (Δ), they propose a phenomenological theory in which a distribution of charging energies exists, due mainly to the randomness in the intergranular resistance rather than in grain diameter. More recently, extraordinarily large magnetoresistance which approaches 100% is found in an ultrathin $La_{0.7}Ca_{0.3}MnO_3$ film by Ziese[10] and the mechanisms are discussed on the network effects and the contribution from magnetic energy term E_m. Clearly these complex properties of magnetotransport in granular manganites could not be understood in a simple spin-polarized tunneling model as for conventional granular ferromagnets, which is well de-

scribed by Inoue and Maekawa.[11] Also the magnetoresistance never obeys the equation[11]

$$MR = \frac{P^2 m^2}{1 + P^2 m^2}, \tag{1}$$

where m is the reduced magnetization of the systems.

Our purpose in this paper is to address the complex magnetotransport phenomena in granular manganites described above and to account for the origin of the enhancement of magnetoresistance found in recent experiments. To do this, a bond-disordered random resistor network is proposed in which the transport process is assumed to be controlled by both the charging energies of grains and spin-polarized tunneling through grain boundaries. The effects of network and band-bending at the grain boundaries on the magnetotransport (including low-field magnetoresistance, high-field magnetoresistance, and Coulomb gap) are investigated. Our results are compared with the recent experiments on nanometric $La_{2/3}Sr_{1/3}MnO_3$ and $La_{2/3}Ca_{1/3}MnO_3$, good agreements are found.

II. EXPRESSION OF INTERGRANULAR CONDUCTANCE G_{ij}

It is well established that the electrical conduction in granular systems results from the transport of electrons or holes from a charged grain to a neutral one. This requires the generation of a charge carrier by removing an electron from a neutral grain and placing it in the neighboring hitherto neutral grain at the expense of electrostatic charging energy $E_c = e^2/2C$, where C is the capacitance of a grain. An additional factor E_c/k_BT is thus introduced to the whole resistance. As a consequence, at low temperatures and for small grains it is very difficult to activate such a mechanism. The electrical transport is effectively blocked.

The typical $\ln R \sim \sqrt{\Delta/T}$ behavior of the resistance in nanometric manganites shows that the electrical transport is similar with that in the conventional granular systems, where the transport is controlled by the combination of the tunneling through an insulating barrier and the charging energies of grains. In the low electrical field regime the charge carriers are thermally activated with the number density $\sim \exp(-E_c/k_BT)$, so the conductance between two grains can be given as[11]

$$G_{ij} = G_{ij}^T \exp(-E_c/k_BT), \tag{2}$$

with G_{ij}^T referring to the tunneling mobility between grains as[11]

$$G_{ij}^T \propto (1 + P^2 \cos\Theta) e^{-2\kappa s}, \tag{3}$$

where

$$P = \frac{D_\uparrow - D_\downarrow}{D_\uparrow + D_\downarrow} \tag{4}$$

and

$$\kappa = \sqrt{2m_e\phi/\hbar^2}. \tag{5}$$

Here, Θ is the relative angle between the orientation of the magnetization of two grains; $D_\alpha(\alpha = \uparrow, \downarrow)$ is the density of states at the Fermi energy E_F for electrons with spin α; P is the spin polarization; and s, m_e and ϕ are the thickness of the tunneling barrier, the effective mass of electrons and the barrier height, respectively.

Unlike conventional granular ferromagnets, grain boundaries in manganites are the regions of magnetic disorders. The effects of such disorders on the magnetotransport have been discussed in many ways, e.g., Evetts *et al.* treated magnetoresistance in bicrystal phenomenologically in terms of changes in local magnetic order[12] and Lee *et al.* assumed the grain boundaries as paramagnetic impurities in their second-order tunneling model.[6] Here in this paper, by the analogy of the study in a high-temperature superconducting cuprate, the magnetic origin of ϕ can be understood by introducing the concept of band-bending effects at the grain boundaries.[13,14] Furukawa has shown that for colossal magnetoresistance materials the magnetization M induces a shift in the chemical potential with $\Delta\phi \sim M^2$ (Ref. 15). As a result of the dislocations and defects, there is a suppressed magnetic order close to the grain boundaries. In nanometric manganites, the grain boundary is supposed to be paramagnetic.[12,13] The energy barrier comes from the difference in chemical potential in the grain boundaries as compared to the grain bulk.

The height of the built-in potential (ϕ) can be expressed as[13,14]

$$\phi \propto \Delta\phi_B - \Delta\phi_{GB} \propto M_B^2 - M_{GB}^2, \tag{6}$$

where M_B and M_{GB} are the magnetization of grain bulk and grain boundary, respectively.

For the ferromagnetism of bulk, the reduced magnetization m_B can be expressed with a mean-field self-consistency equation,

$$m_B(h,t) = B_J\left(\frac{3J}{J+1}\frac{m_B + h}{t}\right), \tag{7}$$

where B_J is the Brillouin function defined by

$$B_J(x) = \frac{2J+1}{2J}\coth\left(\frac{(2J+1)x}{2J}\right) - \frac{1}{2J}\coth\left(\frac{x}{2J}\right), \tag{8}$$

where J is the angular momentum quantum number, $t(T/T_c)$ is the normalized temperature, $h(H/H_e)$ is the normalized magnetic field and H_e is the saturation exchange field. For the optimal perovskite manganites of the form $A_{2/3}B_{1/3}MnO_3$, J is assumed to be 1.83.

For the paramagnetism of grain boundaries, the reduced magnetization m_{GB} is expressed via the Langevin function,

$$m_{GB}(h,t) = \coth(\mu H/k_BT) - k_BT/\mu H, \tag{9}$$

where μ is the effective magnetic moment of isolated Mn spins or their clusters.

Thus the built-in potential can be expressed as

$$\phi = \phi_0(m_B^2 - m_{GB}^2), \tag{10}$$

where ϕ_0 is barrier height when $m_{GB}=0$. Here we assume that the grain boundaries possess almost the same saturation magnetization with grain bulk.

We can find from Eq. (10) that the tunneling barrier disappears when the temperature approaches the Curie tempera-

ture T_c. At low temperatures, because the $\Delta\phi_B$ is insensitive to external magnetic field, which is already saturated for its ferromagnetism, while the $\Delta\phi_{GB}$ increases linearly with an external magnetic field, the barrier height is thus effectively decreased under an external high field. This kind of change from the temperature above T_c to low T is thought to be the origin of appearance of intergranular resistance and continuous decrease of resistance at high external magnetic field in bicrystalline manganites.[13,14]

From the above consideration, we can find the external magnetic field enters G_{ij} not only in the variable of Θ as in conventional granular systems, but also in the barrier height ϕ.

III. FRAMEWORK OF EFFECTIVE-MEDIUM THEORY

The resistor network model has become one of the most extensively studied models in disordered systems. Here we consider a bond-disordered random resistor network on a hyper cubic lattice of dimension $d'>1$ ($d'=2$ for 2D, $d'=3$ for 3D, etc.). The effective medium theory gives a reasonable description of the transport properties in a resistor network.[16] This approach is old and had been devised for the transport properties of inhomogeneous materials first by Bruggeman and then independently by Landauer. Its successful application of the percolation theory has drawn the attention of many others in this field. Consider a random resistor network by a homogeneous effective network or an effective medium where each bond has the same average or effective G_e. The value of G_e is calculated in a self-consistent manner. To accomplish this, one bond embedded in the effective medium is assigned the conductance distribution of the actual random network. The value of G_e is then determined with the condition that the voltage fluctuation across the special bond within the effective medium, when averaged over the proper conductance distribution, is zero. The voltage (v) developed across such a special bond can be calculated for a discrete lattice of $2d'$ nearest neighboring as[16]

$$v = \frac{G_{ij} - G_e}{G_{ij} + (d'-1)G_e}. \tag{11}$$

For a bond-disordered random resistor network, the effective-medium theory condition described above, i.e., $\langle v\rangle = 0$, now reads as

$$\left\langle \frac{G_{ij} - G_e}{G_{ij} + (d'-1)G_e} \right\rangle = 0. \tag{12}$$

The above equation can be rewritten as

$$\int \frac{f(G)dG}{G + (d'-1)G_e} = \frac{1}{d'G_e}, \tag{13}$$

where $f(G)$ is the distribution of intergranular conductance.

Sheng has pointed out that in order to ensure the homogeneity of the metallic grain concentration, the ratio D/s should have the same value for the different regions although both D (grain diameter) and s (intergranular distance) may have a wider distribution.[17] Follows that the product of sE_c is invariant for a certain system, which can be written as[17]

$$sE_c = c, \tag{14}$$

in which c is a constant.

With the condition of $sE_c=c$, there exists the tunneling distance

$$s_m = \sqrt{\frac{c}{2\kappa k_B T}}, \tag{15}$$

which makes the intergranular conductance maximum. Thus we can find the dominant contribution to conductance at high temperature is due to the tunneling between small grains (large E_c) separated by thin tunneling barriers (small s), while at low temperatures the dominant contribution is due to large grains (small E_c) separated by thick tunneling barriers (large s). In the steepest descent approximation, the $G_{ij}(s=s_m)$ is selected to represent the effective conductance of the whole systems. For the magnetotransport in the model by Inoue and Maekawa, the steepest descent approximation is used and the averages over the exponential factor and the pre-exponential factor are conducted separately.[11] This might be proper for the conventional granular ferromagnets, where the spin-polarization is small and at the same time there is a wide distribution of intergranular distance. However, for the high degree of spin-polarization and the relatively narrow distribution of intergranular distance, the contribution from the non-maximum intergranular conductances in the exponential factor should not be neglected.

In order to account for the properties described above and the fact of exponential decrease of conductance when the tunneling distance is away from s_m, we suppose

$$P(s) = \frac{s}{s_m^2}\exp(-s/s_m), \tag{16}$$

to mimic the distribution of intergranular conductance in the network, which is peaked around s_m.

The distribution of intergranular conductance is determined by both $P(s)$ and $f(\Theta)$. If we assume all the grains are superparamagnetism and have the same effective magnetic moment μ as in Ref. 18, the effective conductance can be obtained by

$$\frac{1}{d'G_e} = \frac{1}{(2\pi\Omega)^2}\int_0^\infty P(s)ds\int_{-1}^1 d\cos\theta_1\int_{-1}^1 d\cos\theta_2\int_0^{2\pi} d\varphi_1 \tag{17}$$

$$\times\int_0^{2\pi} d\varphi_2 \frac{e^{h(\cos\theta_1+\cos\theta_2)}}{G_0(1+P^2\cos\Theta)e^{(-c/sk_BT-2\kappa s)} + (d'-1)G_e}, \tag{18}$$

where

$$\cos\Theta = \cos\theta_1\cos\theta_2 + \sin\theta_1\sin\theta_2\cos(\varphi_1-\varphi_2), \tag{19}$$

$$\Omega = \int_o^{\pi} \sin\theta\, d\theta\, e^{h\cos\theta}, \tag{20}$$

$$h = \mu H/k_B T. \tag{21}$$

Generally speaking, the grains are ferromagnetic in nanometric manganites. Here we focus on the absolute value of the magnetoresistance from two extreme states. In the demagnetized (DM) state, the effective conductance can be obtained from

$$0.5 \times \int_0^{\infty} P(s)ds \int_0^{\pi} \frac{\sin\theta\, d\theta}{G_0(1+P^2\cos\theta)\exp(-c/sk_BT - 2\kappa s) + (d'-1)G_e(DM)} = \frac{1}{d'G_e(DM)}. \tag{22}$$

In the magnetized (M) state (when $H=H_s$),

$$\int_0^{\infty} \frac{P(s)ds}{G_0(1+P^2)\exp(-c/sk_BT - 2\kappa s) + (d'-1)G_e(M)} = \frac{1}{d'G_e(M)}. \tag{23}$$

The LFMR is defined as the relative difference between these two states, which results from the rotation of magnetization of neighboring grains under an external magnetic field:

$$\mathrm{LFMR} = \frac{G_e(M) - G_e(DM)}{G_e(M)}. \tag{24}$$

The field needed to rotate the grain in nanometric manganites is usually less than $0.5T$, thus in such a small field, the barrier height is not affected.

It is interesting to compare our results with those from models of resistors in parallel and resistors in series. The quantities for comparison are described briefly below.

The conductance of the resistor in parallel is given as[11]

$$G = G_0 \int\int ds\, d\theta f(s)g(\Theta)(1+P^2\cos\Theta) \times \exp(-2\kappa s - E_c/k_BT), \tag{25}$$

where $f(s)$ and $g(\Theta)$ are, respectively, the distribution functions of s and Θ.

With the steepest descent approximation and $\langle\cos\Theta\rangle = m^2$, the conductance can be written as[17]

$$G = G_0(1+P^2m^2)\exp(-\sqrt{8\kappa c/k_BT}), \tag{26}$$

where m is the relative magnetization of the system. It is also noted that the experimental observed Coulomb gap Δ is proportional to $8\kappa c/k_B$.

So the expression for the magnetoresistance is[11]

$$MR = \frac{P^2m^2}{1+P^2m^2}, \tag{27}$$

and saturated value is $P^2/(1+P^2)$.

It is argued that for the main conduction path is determined by the exponential factor $-2\kappa s$ in the tunneling conductance, the magnetoresistance is determined by one-dimensional resistor network. The quantity from $\langle R(\theta)\rangle$ for the resistors in series is[10]

$$\langle R(H=0)\rangle \propto 0.5 \times \int_0^{\pi} \frac{\sin\theta\, d\theta}{1+P^2\cos\theta} = P^{-2}\,\mathrm{arctanh}(P^2) \tag{28}$$

and

$$\langle R(H=H_s)\rangle \propto \frac{1}{1+P^2}. \tag{29}$$

Thus

$$MR = \frac{\langle R(H=0)\rangle - \langle R(H=H_s)\rangle}{\langle R(H=0)\rangle} = \frac{(1+P^2)P^2\,\mathrm{arctanh}(P^2) - 1}{(1+P^2)P^2\,\mathrm{arctanh}(P^2)}. \tag{30}$$

In fact, the difference between these resistor network models are often overlooked in the practical application and results are confusing. In the following we will discuss them in detail.

IV. RESULTS AND DISCUSSIONS

First we will show the network effects on LFMR. In Fig. 1, we have plotted the P dependence of LFMR from Eq. (24) for 2D and 3D cases and compared them with the results from Eq. (27) and Eq. (30). It is found that when P is less than 0.5 (the value of normal ferromagnets), these models give almost the same LFMR. However, when P increases beyond 0.5, a significant difference can be seen among these results. The LFMR calculated from Eq. (24) for a 3D case with $P=1$ is 56%, which is the exact value of LFMR in half-metallic CrO_2 powder compacts in the experiment by Coey *et al.*[4] As for Eq. (30), for the reason of all the resistor in series, a single insulator (in this case when $\theta=\pi$) can block the whole current path. Thus in the demagnetized state, the resistance can be infinite, while the resistance is finite in a magnetized state, and so the magnetoresistance approaches 100%. This can be regarded as the upper bound of LFMR in half-metallic granular systems. In real systems, the current can percolate above the percolation threshold. The higher

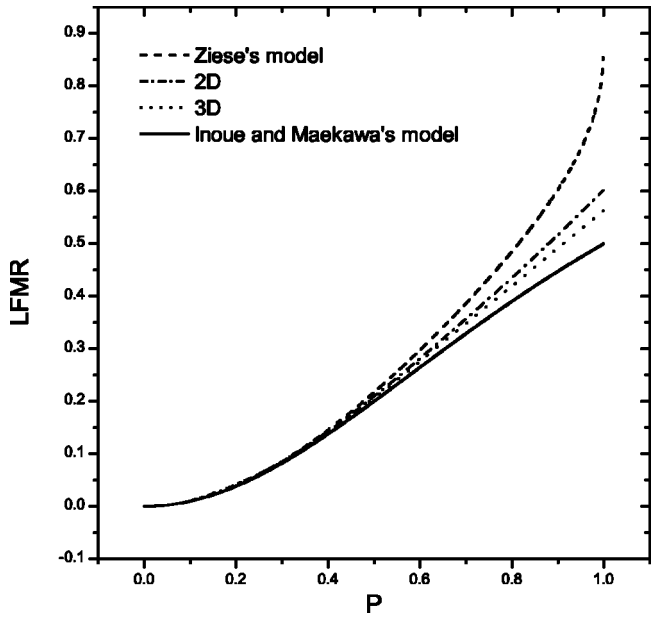

FIG. 1. P dependence of LFMR for three models.

dimension of the systems, the easier for the current to percolate. While for the model of resistor in parallel, the MR is underestimated.

Unlike the conventional granular metals, the surface layer of manganite grains is high resistive and insulating. The grains consist of a metallic core with diameter D and an insulating shell with thickness t. And

$$\Phi = D + 2t, \tag{31}$$

where Φ is the grain size defined in Ref. 7. A low limit of the thickness t can be obtained assuming that it has a magnetization $M_S=0$. With this assumption, in the core-shell model,[7]

$$t = (\Phi/2)(1 - (M_S(\Phi)/M_S(bulk))^{1/3}). \tag{32}$$

So the dependence of t on Φ can be obtained. It is found that t itself increases with the decrease of Φ.[10] When the grain size (Φ) is changed, both D and $s(=2t)$ vary and they vary in an opposite way, i.e., D decreases with the decrease of Φ and s increases with the decrease of Φ. The variation of D/s $\times(=c)$ has the same trend as Φ. Thus with the decrease of grain size, the value of c monotonically increases. In the calculation above and below, we have chosen $\kappa=1$ nm^{-1} and c is in units of $k_B\cdot$nm and make qualitative comparisons with experimental data on both the Coulomb gap and magnetoresistance. Since $E_C=e^2/8\pi\epsilon_0\epsilon(=10)r$, when $c=10$, it means r is around 50 nm and at the same time s is around 1 nm.[7–9]

Since the incorporation of elements of grain diameter (D) and intergranular distance (s), and their distributions through $P(s)$ in the resistor network, it is interesting to investigate their influence on conductance and magnetoresistance. In Fig. 2(a), we have studied the temperature dependence of conductance. The G_e/G_0 is plotted versus $T^{-1/2}$. The linear dependence agrees with the experimental observations. Clearly, the larger c, the smaller the conductance. In Fig. 2(b), we have shown the temperature dependence of conductance under an external magnetic field of $8T$. We can see the conductance still obeys the $T^{-1/2}$ rule, however the Coulomb gap (Δ) (which is magnetic field insensitive in conventional granular systems) is lowered and the resistance is effectively decreased. To have a quantitative picture between an external magnetic field and a Coulomb gap (Δ), in Fig. 2(c) we have $1/c$ dependence of the Coulomb gap (Δ) which is obtained from the linear fit of the temperature dependence of conductance both at $H=8T$ and $H=0T$ from 5 K to 100 K (the linear fitting process is not shown here). We can find, besides the effective decrease of Δ upon the application of external magnetic field, the variation of it increases with the increase of c, i.e., the smaller the grain size, the stronger the dependence of the Coulomb gap (Δ) on the field. This serves as the explanation of experimental observation by Garcia-Herandez *et al.*,[9] where the field dependent Coulomb gap could not be readily understood in the picture of transport in conventional granular systems. The experimental data are shown in the inset for comparison. In Fig. 2(d), we have plotted the field dependence of the Coulomb gap (Δ), where the value of Δ at each field is obtained, as described above. The experimental data on the smallest granular $La_{2/3}Ca_{1/3}MnO_3$ are reproduced in the inset. We can find our results agree with experimental data well. It is noted that a phenomenological model based on the consideration of intergranular connectivity was proposed by Garcia-Herandez *et al.*[9] for this problem, but with four fitting parameters included. We can find the effect of intergranular connectivity and its variation with an external magnetic field have been considered explicitly in Eq. (10). Moreover, since our calculation of the Coulomb gap is performed on a complete resistor network, the relationship between the Coulomb gap and magnetoresistance is also revealed.

In Fig. 3(a), we have shown the $1/c$ dependence of LFMR. It is found that LFMR depends weakly on the values of c. Our calculations agree with experimental observations in Ref. 7, which is shown in the inset of Fig. 3(a). In the nanometric $La_{2/3}Ca_{1/3}MnO_3$, the LFMR is almost grain size independent. It should be noted that the increase of LFMR with a decrease of grain size in the μm scale is due to the increase of weak-link grain boundaries, i.e., the increase of effective tunneling barriers. Here we discuss the cases where all the grains are well separated by insulating grain boundaries.

By now, judging from the calculated low-field magnetoresistance and the temperature dependence of resistance, we can find that the dominant contribution in the network is due to the grains with $s=s_m$. For the distribution form of Eq. (16) has guaranteed that the fraction of conductors near $G_{\max}(s=s_m)$ is larger than the percolation threshold (p_c), the essence of the problem has been grasped in our approach.

As described above, in granular manganites the field dependence of magnetoresistance is usually divided into two parts. Following a rapid decrease of resistance at fields around $0.5T$ (LFMR), there is an almost linear term.

In the two-channel model, the MR is written as[8]

$$R(H)/R(0) = 1/[1 + aM^2(H)/M_s^2][1 + bH], \tag{33}$$

where a and b are microstructure dependent parameters.

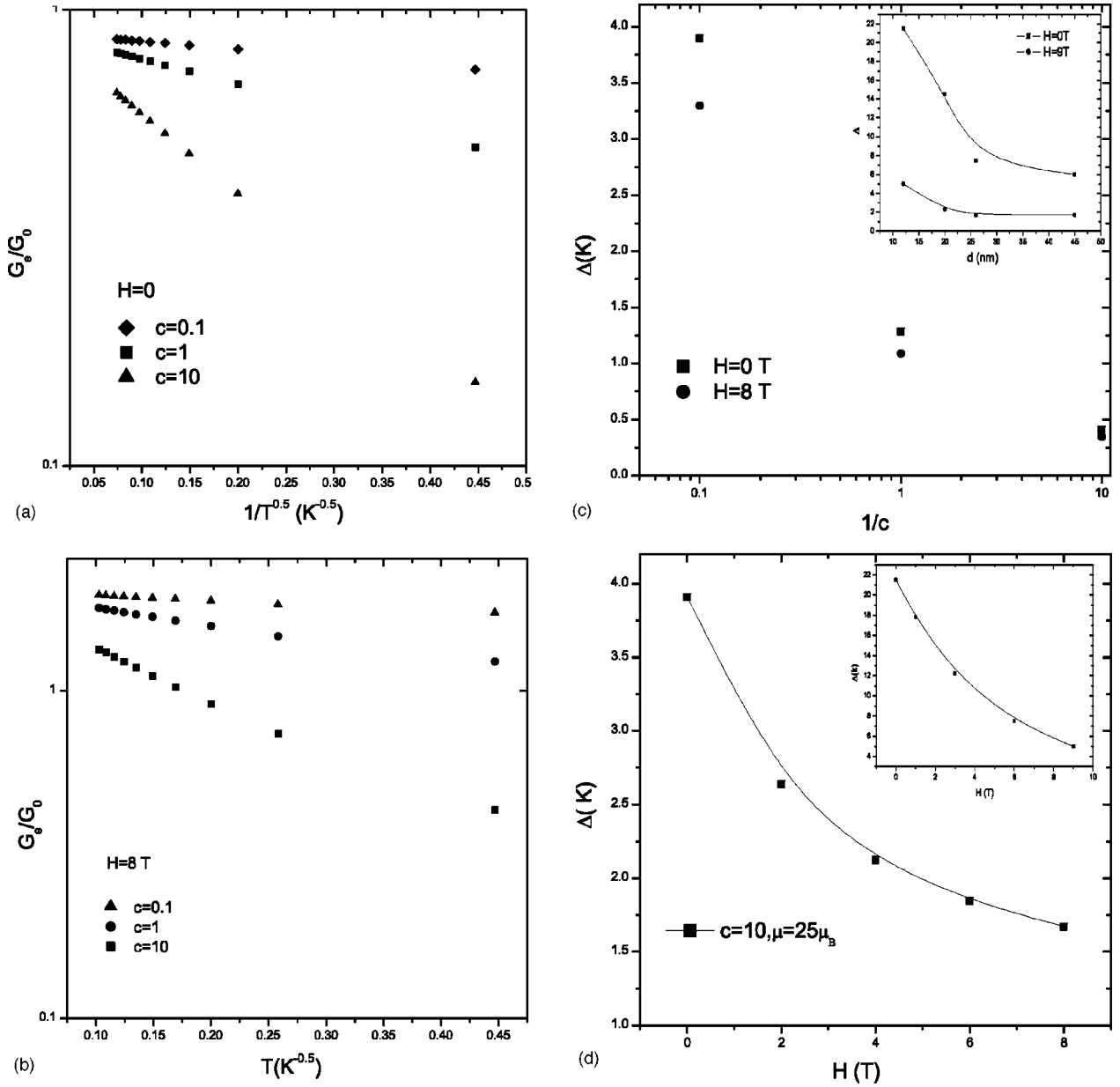

FIG. 2. (a) The temperature dependence of zero field conductance for samples with $c=10$, 1 and 0.1. (b) The temperature dependence of conductance for samples with $c=10$, 1, and 0.1 at a field of $H=8T$. (c) The $1/c$ dependence of the Coulomb gap (Δ) for a zero field case and $H=8T$. The inset shows the experimental data from Ref. 9. (d) The field dependence of the Coulomb gap (Δ) for a sample with $c=10$. The inset shows the experimental data from Ref. 9.

It is also expressed in the form of magnetoconductance by Lee *et al.*,[6]

$$\sigma/\sigma_0 = a' + b'H, \tag{34}$$

where a' and b' are parameters.

Here we focus on the high-field response which is due to the evolution of the band structure at grain boundaries. The HFMR is defined as

$$\text{HFMR} = d(G(H > H_s)/G(H = H_s))/dH. \tag{35}$$

In Fig. 3(b), we have plotted the field dependence of $G_e/G(H=H_S)$, which represents the high field response, irrelevant to the rotation of the magnetization of grains. It is found that there is a linear increase of conductance as a function of the field. It should be noted that our results also indicate that conductance varies linearly with a magnetic field, rather than resistance. This kind of observation has been emphasized in the experiments by Lee *et al.* on a series of manganites.[6] In Fig. 3(c), we have shown the $1/c$ dependence of HFMR at 5 K. As mentioned above, c monotonically increases with the decrease of grain size. Thus our results indicate HFMR increases with the decrease of grain size. This kind of variation has also been found in Ref. 7, which are shown in the inset.

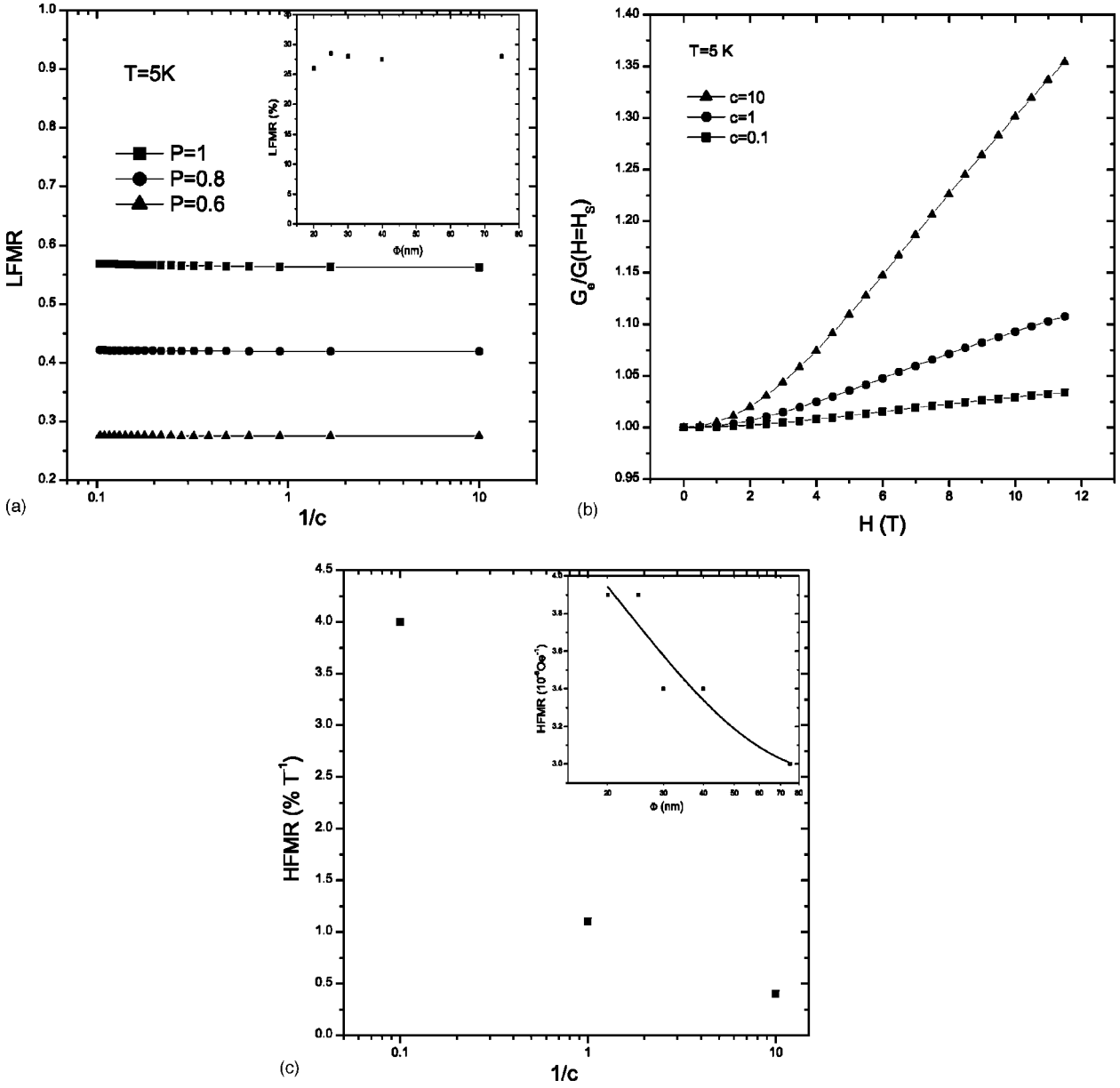

FIG. 3. (a) The $1/c$ dependence of LFMR at 5 K for $P=1$, 0.8 and 0.6. The inset shows the experimental data on the grain size (Φ) dependence of LFMR from Ref. 7. (b) The field dependence of $G_e/G(H=H_S)$ at 5 K for samples with $c=10$, 1 and 0.1. (c) The $1/c$ dependence of HFMR at 5 K. The inset shows the experimental data (square dots) on the grain size (Φ) dependence of HFMR in Ref. 7. The solid lines are guides for the eyes.

From the previous consideration of the field dependence of magnetoresistance and its evolution with grain size, we can conclude the great enhancement of magnetoresistance observed in some granular manganites[10] with an ultrafine grain size is not directly related with the high value of spin-polarization, but is due to the effective decrease of barrier height in the sample where the Coulomb gap (Δ) is giant. At the same time, we would like to point out that in this article we focus our study on the temperatures below Curie temperature T_c. Near T_c, the intrinsic phase-separation between the metallic phase and insulating one play an important role in determining the magnetotransport in polycrystalline manganites.[19]

V. SUMMARY

In summary, we have studied the magnetotransport in the nanometric manganites within a bond-disordered resistor network. The transport process here is assumed to be controlled by both the charging energies and spin-polarized tunneling through grain boundaries. The effects of network and band-bending at the grain boundaries on the magnetotransport (LFMR, HFMR and Coulomb gap) are investigated. Our results on the grain size dependence of magnetotransport agree with experiments very well and the origin of recently found extraordinarily large magnetoresistance is well explained.

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