Monte Carlo simulation of tunneling magnetoresistance in nanostructured materials

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A two-level model of tunneling magnetoresistance (*MR*) containing coplay of the relative orientation of moments between clusters, the intrinsic characters of clusters, and the Coulomb blockade effect, is presented. Based on the Monte Carlo simulation, spin configurations, *MR* and resistivity as function of temperature and field were studied, respectively. Simulated results are found to be in good agreement with the recent experimental data. The unusual enhancement of *MR* is attributed to the interaction between the clusters and the intrinsic character of clusters. Although the Coulomb blockade effect plays little role on *MR*, it has an important effect on the resistivity at low temperature.

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I. INTRODUCTION

Tunneling of spin-polarized electrons and magnetoresistance in complex phase systems are of great current interests for both the discovery of new phenomena and their potential applications in high-density memory devices. Tunneling magnetoresistance $[MR=(R(0)-R(H))/R(0)]$ was first found by Julliere[1] in 1975. Recently, large MR ratios have been observed in tunneltype nanostructures, e.g., MR ratio of 46% in $Co/Al_2O_3/CoFe$ tunnel junctions,[2] MR' $[MR'=(R(0)-R(H))/R(H)]$ as large as 158% at $T=300$ K in polycrystalline $Zn_{0.41}Fe_{2.59}O_4$ with grains coated by α-Fe_2O_3,[3] MR' exceeding 500% in half-metallic Fe_3O_4 nanocontacts[4] and high-MR ratios in other materials.[5–10] In order to explain these large MR ratios, especially their dependence upon temperature and field, a model involving the interplay between spin-dependent tunneling and Coulomb blockade has been used.[3,5–8,11–13] However, Holdenried *et al.*[9] pointed out that the above interplay model (Mitani model[8]) is probably not the fundamental one, and introduced a different simple model considering the misalignment of magnetic moments at the surface of cluster. Meanwhile, from the measurements of Schelp *et al.*[6] it is impossible to conclude that the Coulomb blockade contributes to the temperature dependence of *MR*. Therefore, although several authors have suggested the theoretical models with single domain cluster or electrode for tunneling MR,[11–16] it is still an opening question for the origin of large *MR* ratio at high temperature!

As we know, the properties of materials consisting of magnetic clusters should be determined by both the intrinsic properties of the clusters and the interactions between clusters. Obviously, it is an oversimplification that the clusters or the electrodes are treated as a single domain system with the same basic spin orientation. Recently, attention has been paid to the intrinsic properties of clusters,[3,9,17–19] and, especially, to the surface spin disorder and interface magnetic anisotropy. Also, Coey *et al.*[5] suggested that the change of MR under an applied field be possibly due to the reduction of the effective height or width of the tunnel barriers and the polarization of disordered spins in the interface. In this paper, based on our recent experimental results[3] and the standard Monte Carlo simulation, a two-level tunneling MR model containing coplay of the relative orientation of moments between clusters, the intrinsic character of clusters, and the Coulomb blockade effect, is presented. Effects of temperature and magnetic field on resistivity and MR are studied systematically. It has been found that large *MR* ratio at high temperature results mainly from the intrinsic properties of clusters.

II. MODEL

Julliere suggested a classical theory for tunneling based on conduction-electron spin-polarization values (P_1 and P_2) of the electrodes, and obtained the tunneling *MR* as $MR=2P_1P_2/(1+P_1P_2)$.[1] Inoue and Maekawa expressed the tunneling conductance G as[15]

$$G=\frac{e^2}{h}|T|^2\propto(1+P_t^2\cos\theta)e^{-2\varkappa S_0}, \tag{1}$$

where $|T|^2$ is the transmission coefficient, θ is the angle between the moments of the two ferromagnets, P_t is the spin polarization, S_0 is the thickness of the barrier, $\varkappa$ is $\sqrt{2m^*(U-E_F)/\hbar^2}$ with the Fermi energy E_F, the effective mass of electrons m^*, and the barrier height U, as shown in Fig. 1(a). When the cluster is treated as a single domain, the thickness of the magnetic barrier S_m at surface is taken as zero. Equation (1) can be approximately rewritten as the expression of resistivity,

$$\rho(H,T)\propto\int\frac{e^{2\varkappa S_0}}{(1+P_t^2\cos\theta)}g(\theta)d\theta\approx P_t^2(p-\langle\cos\theta\rangle)e^{2\varkappa S_0} \tag{2}$$

with $p=1/P_t^2$.

In nanostructured materials, the tunneling of an electron into a small particle or cluster gives rise to an interesting effect, namely the Coulomb blockade.[6,8,11] This effect brings a charging (Coulomb) energy of $E_c=e^2/2C$ with C for the

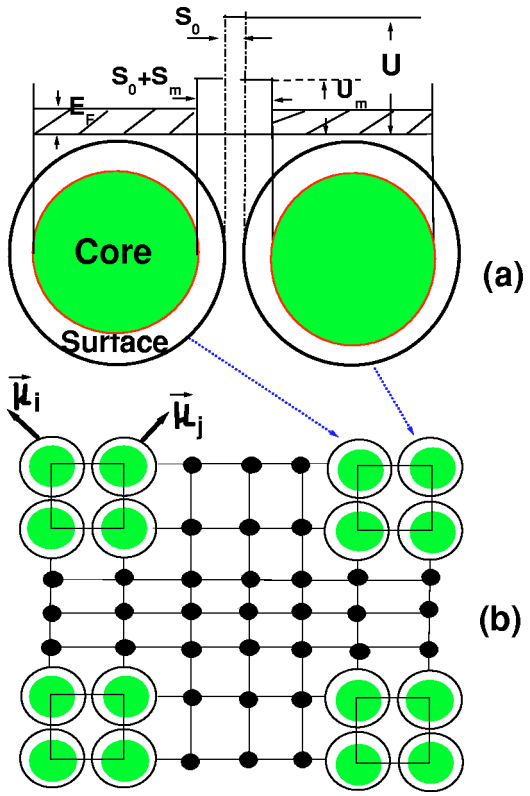

FIG. 1. (a) Schematic illustration of spin-tunneling effect in the clusters containing complex spin configurations; (b) the sketch of the clusters located on a net.

capacitance of the cluster. Considering the Coulomb blockade effect, the resistivity can be expressed by

$$\rho(H,T)\propto(p-\langle\cos\theta\rangle)e^{2\varkappa S_0+E_c/2k_BT}. \tag{3}$$

The tunneling of electron between the clusters containing complex spin configurations is depicted in Fig. 1(a). The ground state of the magnetic clusters is strongly influenced by finite size and microstructural details of both the core and surface. Two-level interaction model is established. At first level, we model the system as a collection of three-dimensional nanosized spherical clusters (single domain), with each cluster located on a lattice site of a simple cubic multilayered net containing $10\times10\times5$ cells with the distance r_0 between both nearest-neighbor clusters,[16] as seen in Fig. 1(b). It is assumed that the direction of the uniaxial anisotropy axis is taken to be random in space. The system Hamiltonian containing the classical dipolar interaction, anisotropy, and Zeeman energies is given by

$$E=K\left\{D\sum_{(ij)}\left[\frac{\vec{\mu}_i\cdot\vec{\mu}_j}{r_{ij}^3}-\frac{3(\vec{\mu}_i\cdot\vec{u}_{ij})(\vec{\mu}_j\cdot\vec{u}_{ij})}{r_{ij}^3}\right]-\sum_i(\vec{\mu}_i\cdot\vec{u}_i)^2-H'\sum_i\mu_i^z\right\}, \tag{4}$$

where D is the reduced magnitude of the dipolar coupling, $\vec{u}_{ij}$ is the unit vector for the vector connecting two moments unit vector $\vec{\mu}_i$ and $\vec{\mu}_j$ at site i and j, respectively, K is the single-site uniaxial anisotropy energy (with assumption that the K value at every site is same), $\vec{u}_i$ is the unit vector of easy axis at the site i, H' is μ_0H_e/K with H_e for the external magnetic field along the z axis and μ_0 for the magnetic moment. $\Sigma_{(ij)}$ is performed over the dipolar interactions between moments with a cutoff at a distance $r=c_t$ in units of nearest-neighbor distance r_0. In our calculation c_t is taken to be 6.

At second level, based on the fact that, as high-resolution TEM image clearly indicated, the two-phase system has the structure of the $Zn_{0.41}Fe_{2.59}O_4$ grains coated by an α-Fe_2O_3 grain boundary,[3] the cluster can be divided into surface and core with different interactions. Thus, a magnetic barrier appears at the surface. Now, we focus on the change of barrier height U and Fermi energy E_F with the spin configurations of the clusters. The barrier is divided into two parts, U and U_m. It is assumed that U is independent on the magnetic state. While U_m, which results from the surface of clusters with disorder spins or antiferromagnetic coupling, is closely related to the finite size of cluster, the spin configurations, and the microstructure of both the core and surface. The changes of magnetic barrier, including its height and tunneling changes under the external magnetic field have been suggested in $La_{0.7}Ca_{0.3}MnO_3$, EuS, and EuSe.[10,20–22]

According to the tunnel scattering quantum theory,[14,17] in the spin-polarized free-electron approximation, the Hamiltonian of electrons in the core and surface of cluster can be expressed by

$$H_c=\begin{cases}-\dfrac{\hbar^2}{2m}(d/d\xi)^2+U(\xi) & \\ \quad \text{for } \textit{nomagnetic barrier} & \\ -\dfrac{\hbar^2}{2m}(d/d\xi)^2-\vec{h}_b(\xi)\cdot\vec{S}_{i'} & \text{for } \textit{core} \\ -\dfrac{\hbar^2}{2m}(d/d\xi)^2+U_s(\xi)-\vec{h}_s(\xi)\cdot\vec{S}_{i'} & \text{for } \textit{surface},\end{cases} \tag{5}$$

where $-\hbar^2/2m(d/d\xi)^2$ is kinetic energy, $U_s(\xi)$ and $U(\xi)$ are potential energies in the surface and in the nonmagnetic barrier, respectively, $\vec{h}_b(\xi)$ and $\vec{h}_s(\xi)$ are the sum of the molecular and the external magnetic fields in the core and the surface, respectively. They can be expressed by $\vec{h}(\xi)=\vec{H}_e+\vec{H}_{ex}=\vec{H}_e+\Sigma_{j'}^{nn}J_{i'j'}\vec{S}_{j'}$,[17] with $J_{i'j'}$ for the exchange interaction constant. Thus, the total magnetic energy in the cluster can be described by

$$E_M=-\sum_{\langle i'j'\rangle}J_{i'j'}\vec{S}_{i'}\cdot\vec{S}_{j'}-H\sum_{i'}S_{i'}^z, \tag{6}$$

where $\Sigma_{\langle i'j'\rangle}$ is performed for all the pairs at nearest-neighbor sites i' and j'. In the calculation, it is assumed

that: (1) $J_{i'j'}$ with both the sites i' and j' or one of them locating in the core is taken as J, and J is used as the unit of temperature and energy, (2) $J_{i'j'}$ with the site i' and j' all locating in the surface is taken as JN, representing the reduced exchange interaction at the surface. Thus, the reduced field H in Eq. (6) can be expressed by $H=\mu H_e/J$.

It was suggested that the magnetic energy is proportional to the spin correlation of two spins,[12] or, $E_m \propto [1-\langle \vec{S}_{i'} \cdot \vec{S}_{j'} \rangle / S^2]$. Here it is assumed that every spin has the same magnitude of 1. This relation is also proved in our Monte Carlo simulations. Therefore, we can get $E_m^b = J_b[1-\langle \vec{S}_{bi'} \cdot \vec{S}_{bj'} \rangle]$ and $E_m^s = J_s[1-\langle \vec{S}_{si'} \cdot \vec{S}_{sj'} \rangle]$ with $\langle \vec{S}_{bi'} \cdot \vec{S}_{bj'} \rangle$ and $\langle \vec{S}_{si'} \cdot \vec{S}_{sj'} \rangle$ representing the spin correlations in the core and surface, respectively. From Eqs. (3) and (5), we can derive

$$\rho(H,T) \propto (p-\langle \cos\theta \rangle) e^{2\varkappa_m S_m} e^{2\varkappa S_0 + E_c/2k_BT} \tag{7}$$

where

$$\varkappa_m = \sqrt{2m^*[U_s - E_F + J_s(1-\langle \vec{S}_{si'} \cdot \vec{S}_{sj'} \rangle) - J_b(1-\langle \vec{S}_{bi'} \cdot \vec{S}_{bj'} \rangle)]/\hbar^2},$$

$$\varkappa = \sqrt{2m^*[U - E_F - J_b(1-\langle \vec{S}_{bi'} \cdot \vec{S}_{bj'} \rangle)]/\hbar^2},$$

E_F is the chemical potential, S_m and S_0 are the widths of barriers in magnetic surface and nonmagnetic region, respectively, as shown in Fig. 1(a). For the ferromagnetic and ferrite clusters containing several hundreds to thousands of atoms, the contribution of the spin energy and spin correlation in the core to the barrier heights is less important. The dominant effect results from the change of the spin energy and spin correlation in the surface with complex spin structure. To simplify the discussion, we let $S_0=0$, $J_b=J_s=J_0$, $U_i=(U_s-E_F)/J_0$, and $\varkappa_0=2S_m\sqrt{2m^*J_0/\hbar^2}$. Finally, the resistivity can be expressed by

$$\rho(H,T) \propto \rho_s \cdot \rho_m \cdot \rho_c = (p-\langle \cos\theta \rangle) e^{\varkappa_0 \sqrt{U_i - \langle \vec{S}_{si'} \cdot \vec{S}_{sj'} \rangle + \langle \vec{S}_{bi'} \cdot \vec{S}_{bj'} \rangle}} e^{E_c/2k_BT}, \tag{8}$$

where the first term ρ_s results from the relative orientation between moments of magnetic clusters, the second one ρ_m is associated with the intrinsic characters of clusters, the third one ρ_c is due to the Coulomb blockade effect. In Eq. (8), ρ_s is a spin-dependent term due to the relative orientation between moments of the clusters. While the magnetic barrier height in ρ_m term is the difference of electronic magnetic energies at the magnetic surface and in the core. It depends not on the spin directions (up or down). The similar spin-independent tunnel magnetic barrier has been suggested in the other work [10,20–22]. As a special case of Eq. (8), based on a Ising-like model with surface spins parallel and antiparallel to the spins of the cluster core, we can derive the temperature dependence of MR as, $MR(T)=[1-4f(1-f)]MR(0)/(1-4f(1-f)MR(0))$, here f means the ratio of the number of misaligned spins to that of the total ones at the cluster surface. This is the same as Eq. (2) in Ref. 9 derived with different way. Figure 2 shows the MR as a function of f with $MR(0)=0.1$ and 0.5, respectively. From Fig. 2, it is found that, as the array of the spins at the surface is disordered ($f=0.5$) MR equals to 0, while the magnitude of MR increases with the increase of the order degree of the spins at the surface.

Now, let us simulate the spin (or moment) configurations of three-dimensional net consisting of $10\times10\times5$ cells and fcc magnetic clusters using the standard Monte Carlo–Metropolis (MC).[23,24] The first 10 000 MC steps per spin were discarded for equilibrium and thermal averages were taken with next 10 000 MC steps. In the thermal average process, we started to store the simulated parameter values separated by every 20 MC steps to break correlation between successive configurations. At first, we consider $10\times10\times5$ three-dimensional multilayered net. The single domain clusters are placed in the cells. Periodic boundary conditions are applied in x and y directions. Open boundary conditions are applied in z direction. Second, we consider a fcc structure cluster containing 2123 magnetic atoms with 1505 core ones and 618 surface ones (atoms in the coated layer), and 48 shells consisting of 39 core shells and 9 surface shells. Open boundary conditions are applied in all directions. In the simulation, the coefficients in Eqs. (4) and (6) are taken as $J=8.286$ (meV) and $K=0.1J$.

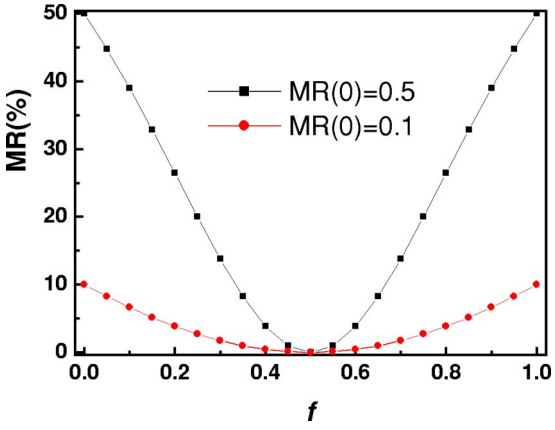

FIG. 2. The $MR(f)$ as a function of f.

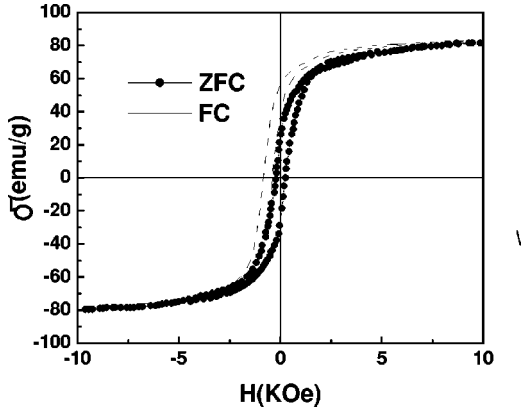

FIG. 3. The magnetic hysteresis loops of zero-field cooled and field-cooled polycrystalline $Zn_{0.41}Fe_{2.59}O_4$ with α-Fe_2O_3 grain boundaries at room temperature.

III. RESULT AND DISCUSSION

Based on our experimental results,[3,25] a polycrystalline grain is considered as a fcc structure cluster. Figure 3 shows the magnetic hysteresis loops of zero-field cooled and field-cooled polycrystalline $Zn_{0.41}Fe_{2.59}O_4$ with α-Fe_2O_3 grain boundaries at room temperature. Prior to the hysteresis loop measurements, the sample were cooled from the Neel temperature of α-Fe_2O_3 to room temperature. The displacement between them strongly proves the existence of antiferromagnetic coupling in grain boundaries. Therefore, the exchange interaction in the surface of clusters is in the form of an antiferromagnetic coupling. In addition, in order to fit our experimental data satisfactorily, different magnitude of *JN* is tried. Finally, it is found that the optimal value for *JN* is -0.05.

Comparing the Curie temperature $T_c=1388$ K and interaction of Co (Ref. 26) with ones of our sample, we set $J_0=9.52$ meV, $S_m=7.0$ (nm). Then we can get $\varkappa_0=7.0$. Additionally, U_i is set for six different values or U_i $(i=0,1,2,3,4,5)=0.0,0.1,0.5,1.0,5.0,10.0$. The spin configurations of the cluster at $T=0.01,0.31,4.21$ are simulated and shown in Figs. 4(a), 4(b), 4(c), respectively. It is found that below Curie temperature the spins in the core retain basically ferromagnetic array, while the spins in the surface are in disorder. The simulated temperature dependences of *MR* and magnetization *M* for U_i $(i=0,1,2,3,4,5)=0.0,0.1,0.5,1.0,5.0,10.0$ and $JN=-0.05$, $H=3.0$ are presented in Fig. 5, respectively. Under external field, the disordered spins in the surface are forced to align along the direction of the field, while parallel spins in the core only have little change. With increasing temperature, the thermal energy causes the spins in the clusters to change their states. However, the extents of the change of the spins in the core and surface are different. The sensitivity difference of the spin array to field and temperature gives rise to the different changes of spin correlation and *MR* with temperature and field.

According to Eq. (8), the *MR* and resistivity resulting from coplay of the relative orientation of moments between clusters, and the intrinsic character of clusters can be obtained. The total *MR*, including those from the cell and intrinsic character of clusters, as a function of temperature for $U_1=0.1$ and $P_t=0.71$ is presented in Fig. 6. Here, the value of P_t is taken from the experimental fact.[25] The temperature dependence of *MR* from cell in Fig. 6 is similar to the simulated result obtained by Xu *et al.*[16] in the three-dimensional multilayer lattice of dipolar interacting fine anisotropy magnetic particles embedded in a nonmagnetic metallic matrix. In addition to the simulated results with Monte Carlo, de-

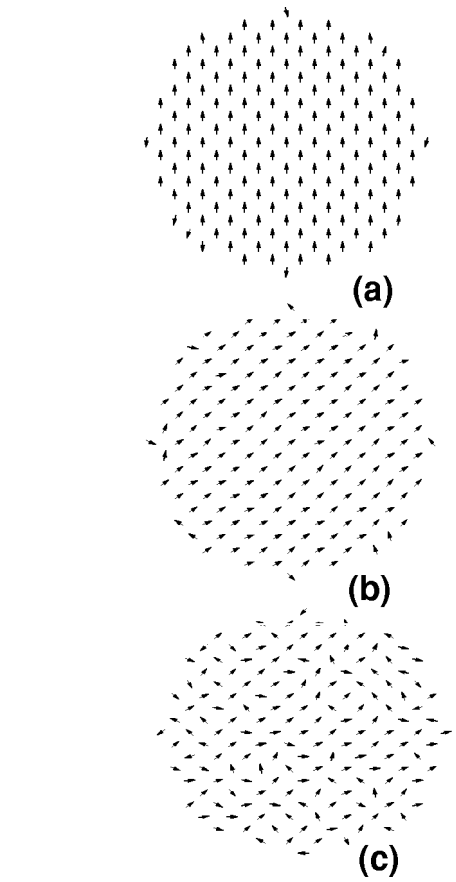

FIG. 4. The spin configuration of cluster with $JN=-0.05$, (a) $T=0.01$, (b) $T=0.31$, and (c) $T=4.21$.

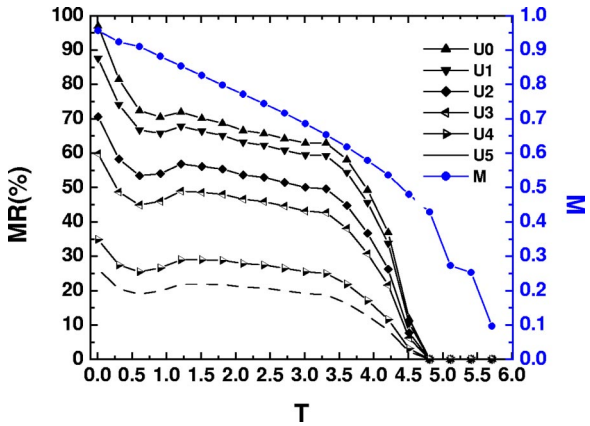

FIG. 5. *MR* and *M* as a function of temperature with U_i $(i=0,1,2,3,4,5)=0.0,0.1,0.5,1.0,5.0,10.0$ and $JN=-0.05$, $H=3.0$.

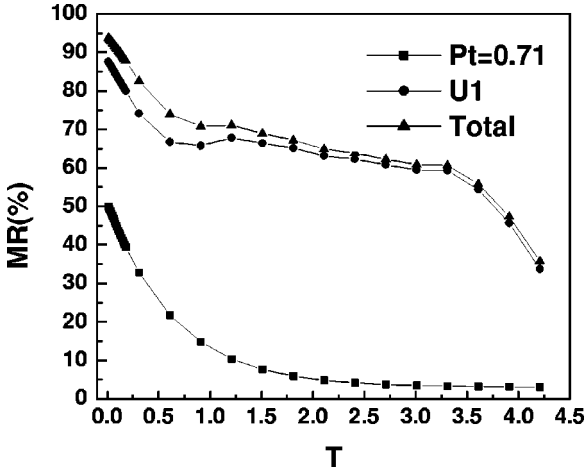

FIG. 6. The temperature dependence of *MR* from cell and intrinsic character of clusters for $U_1=0.1$, $P_t=0.71$.

picted in Fig. 7 are the temperature dependence of MR' from the experiments and fitting with relation $MR'=A+B/T$. Here, we set $3.31J=k_BT_c=k_B\times 318$ ($T_c=318$ K).[3] By comparing the curves, we can find that (1) from the simulation, $MR'=1280\%$ at $T=4.2$ K, and 165% at $T=300$ K. While the experimental magnitudes of MR' are 1280% at 4.2 K and 158% at 300 K, respectively. Therefore, the accordance among the simulated MR' by Monte Carlo, the experimental MR', and the fitted MR' with $1/T$ relation is excellent, which explains experimental fact well; (2) the unusual enhancement of the tunneling MR' is associated with the interaction between the clusters and the intrinsic character of the clusters, and it may not be attributed to the Coulomb blockade effect at low temperature; (3) large MR' ratio at high temperature results mainly from the intrinsic properties of clusters. The various simulated temperature dependence of *MR* has been obtained by choosing different simulation parameters,[27] which is consistent with the recent experiment results.[5,9,13] In addition, we have applied this model [Eq. (8)] to the case in polycrystalline manganites $La_{2/3}(Ca,Sr,Ba)_{1/3}MnO_3$. The simulated magnetoresistance explained the experimental tunneling *MR* at low temperature well.[28]

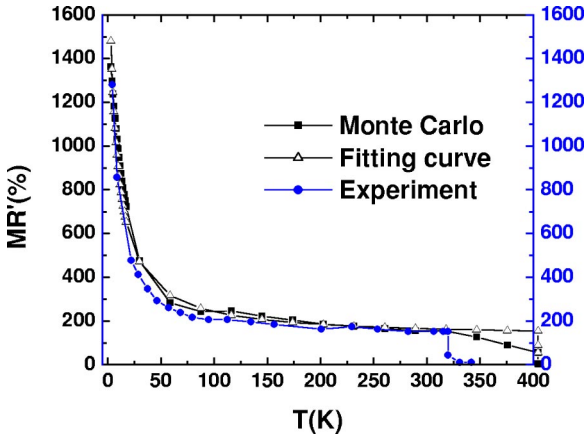

FIG. 7. Simulated and experimental MR′ as a function of temperature with $P_t=0.71$, $JN=-0.05$, $U_1=0.1$.

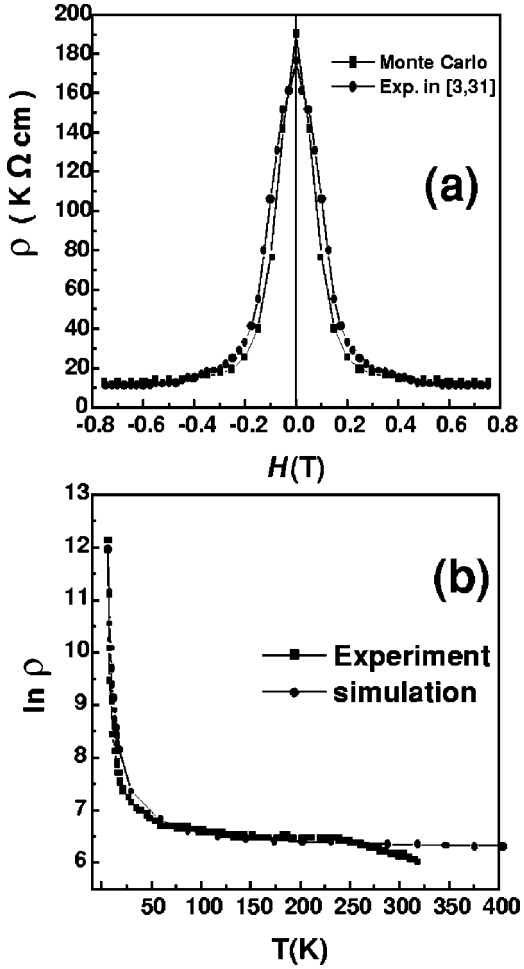

FIG. 8. (a) Simulated and experimental resistivity as a function of field, (b) experimental and simulated resistivity as a function of temperature, with $U_1=0.1$, $P_t=0.71$, $JN=-0.05$, $E_c=0.75$.

Figure 8 (a) shows the field dependence of resistivity for both simulation with $P_t=0.71$, $JN=-0.05$, $U_1=0.1$, and experiment. The accordance between them is obviously demonstrated. However, in simulation, it is found that the experimental result for the temperature dependence of resistivity cannot be explained if only the interaction between the clusters and the intrinsic character of clusters are considered. Therefore, attention should be paid to the Coulomb blockade effect as found in Eq. (8). Figure 8(b) shows the temperature dependence of the logarithm of resistivity from simulation with $P_t=0.71$, $JN=-0.05$, $U_1=0.1$, $E_c=0.75$, and experiment, respectively. By comparing simulated results with experimental ones as shown in Figs. 7 and 8, it is demonstrated that the Coulomb blockade effect does play little role on *MR*, but has an important effect on the resistivity at low temperature, which is consistent with the experimental result of $Co/Al_2O_3/Co$ tunnel junctions in which the Al_2O_3 layer contains a unique layer of small and disconnected cobalt clusters.[6] The results indicate that the resistance is considerably enhanced by the Coulomb blockade effect at low temperatures whereas the relative *MR* ratio changes moderately, which does not support the view point that by changing the energy range of the tunneling the Coulomb

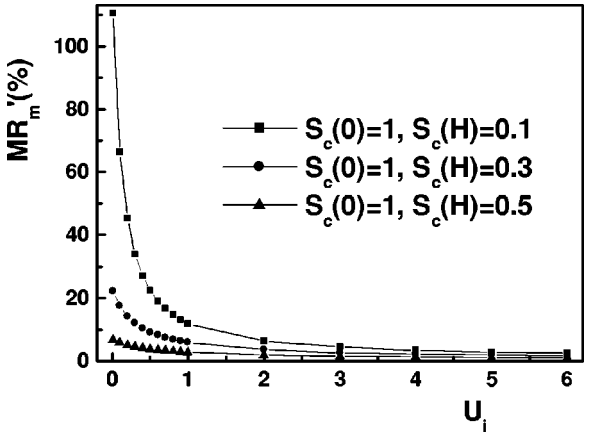

FIG. 9. MR'_m as a function of U_i with $\varkappa_0=7$, $S_c(0)=1$, $S_c(H)=0.1,0.3,0.5$.

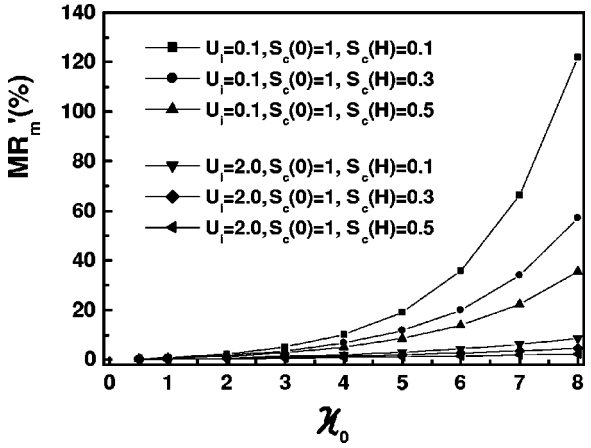

FIG. 10. MR'_m as a function of $\varkappa_0$ with $U_1=0.1$ and 2.0, $S_c(0)=1$, $S_c(H)=0.1,0.3,0.5$.

blockade can result in some enhancement of the *MR* ratio. Therefore, it is thought to be impossible that the Coulomb blockade contributes to the temperature dependence of the *MR*.

Now, we evaluate Eq. (8) and discuss the availability of the model. The first term of Eq. (8) is associated with the relative orientation between moments of magnetic clusters and spin polarization, which has been discussed previously.[15,16] The third one is due to the Coulomb blockade effect, being not related to the magnetic field. Thus, we focus on the second one resulting from the intrinsic characters of clusters. From Eq. (8), we obtain

$$\rho_m = e^{\varkappa_0 \sqrt{U_i - \langle \vec{S}_{si'}(H)\cdot\vec{S}_{sj'}(H)\rangle + \langle \vec{S}_{bi'}(H)\cdot\vec{S}_{bj'}(H)\rangle}}, \qquad (9)$$

where $\langle \vec{S}_{bi'}(H)\cdot\vec{S}_{bj'}(H)\rangle$ and $\langle \vec{S}_{si'}(H)\cdot\vec{S}_{sj'}(H)\rangle$ are correlated with the magnetic field. Let $S_c(H)=\langle \vec{S}_{bi'}(H)\cdot\vec{S}_{bj'}(H)\rangle - \langle \vec{S}_{si'}(H)\cdot\vec{S}_{sj'}(H)\rangle$, $MR'_m=[\rho_m(0)-\rho_m(H)]/\rho_m(H)$. Figure 9 shows the MR'_m as function of U_i with $\varkappa_0=7$, and $S_c(0)=1$, $S_c(H)=0.1,0.3,0.5$, respectively. Figure 10 shows the MR'_m as function of $\varkappa_0$ with $U_i=0.1$ and 2.0, $S_c(0)=1$, $S_c(H)=0.1,0.3,0.5$, respectively. It is found that MR'_m increases dramatically with decreasing U_i and $S_c(H)$, and increasing $\varkappa_0$. From Figs. 9 and 10, it can be seen that small magnitudes of U_i, $S_c(H)$, and large $\varkappa_0$ are the required conditions to enhance the magnetoresistance MR'_m, which means that, first, the potential energies U_s in the magnetic surface barrier should be near the Fermi energy E_F in the core, second, there is J_0 of large enough, third, $S_c(H)$ should be sensitive to the external magnetic field. Therefore, there does not always exist large *MR* in the complex phase systems containing the ferromagnetic or ferrite grain with the antiferromagnetic boundaries.

IV. CONCLUSION

In conclusion, we have introduced a two-level model of *MR* containing coplay of the relative orientation of moments between clusters, the intrinsic character of clusters, and the Coulomb blockade effect. The magnetic cluster is divided into the core and surface consisting of complex spin configurations, which gives rise to the formation of a magnetic barrier. Based on the standard Monte Carlo method, the dependence of *MR* and resistivity on temperature and field is studied, respectively. It is found that the unusual enhancement of *MR* is attributed to the interaction between the clusters and the intrinsic characters of clusters. Large *MR* ratio at high temperature results mainly from the intrinsic properties of clusters. The Coulomb blockade effect does play little role on *MR*, but having an important effect on the resistivity at low temperature.

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