Microscopic simulation of the percolation of manganites

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(Received 17 August 2004; accepted 9 November 2004; published online 30 December 2004)

The one-orbital double exchange model is studied using the METROPOLIS Monte Carlo method and the microscopic resistor network. The phase competition and percolation are displayed microscopically. As far as the resistivity is concerned, the metal–insulator transition is described by the competition between a fraction p of metallic resistors and a fraction $1-p$ of insulating resistors. p can be obtained as a function of temperature T, doping percentage x, and external field H. In the present model, systems with different x, T, and H can be unified into a single class of percolation, which is different from the standard picture. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1848184]

The study of manganites, which exhibit the colossal magnetoresistance effect, is one of the main topics of research within the field of strongly correlated electrons.[1,2] With intermediate doping, the system presents the metal–insulator transition (MIT). In the previous experimental and theoretical works, it is found that, rather than a homogeneous state, the system is characterized by intrinsic inhomogeneity.[1,3] The basic concept underlying is phase separation (PS) and percolation.

In the pioneering work of Mayr *et al.*,[4] the authors studied the resistivity of mixed-phase manganites, using a randomly generated resistor network consisting of a fraction p of metallic resistors (with the resistivity increasing with temperature T) and a fraction $1-p$ of insulating resistors (with the resistivity decreasing with T). With appropriate selection of p, the competition of them could lead to the MIT. Based on it, two further works remain to be done: first, the fraction of metallic resistors needs to be determined (see discussions in, e.g., Ref. 5); and second, as suggested by Burgy *et al.*,[6] the standard percolation (SP) used in Ref. 4 might be different from reality.[7] It should be noticed that the disorder that induces mesoscopic (μm scale) PS and percolation[1] is not randomly arranged, due to, for example, the cooperative nature of the Mn-oxide lattice distortions.[8]

Here we use the METROPOLIS Monte Carlo (MC) simulation of the one-orbital double exchange (DE) lattice model to discuss the above-mentioned topics. The one-orbital DE model can display a significant part of the interesting physics, and remains under theoretical investigation for many years.[1,2,9] The e_g and t_{2g} electrons, though they are fermions, are assumed to have reduced Ising-type spins, ±1, for simplicity. The model Hamiltonian

$$H = -t\sum_{\langle i,j\rangle}(c_{i\sigma}^{+}c_{j\sigma} + \text{H.c.}) - J_H\sum_i s_i S_i + J_{\text{AF}}\sum_{\langle i,j\rangle} S_i S_j - H\sum_i S_i, \tag{1}$$

where $c_{i\sigma}$ and s_i refer to hopping e_g spins, S_i refers to localized t_{2g} spins, $\Sigma_{\langle i,j\rangle}$ stands for nearest neighbor (NN) summation, t is the NN hopping amplitude for the e_g electrons, and the last two terms account for the antiferromagnetic (AF) superexchange interaction and the application of external field, respectively. Here t and k_B are chosen to be unity, and the other parameters T, J_{AF}, and H are all measured in units of t. We suppose $0 < J_{\text{AF}} \ll 1$ (in the model $J_{\text{AF}} = 0.0625$), for the AF superexchange interaction is much weaker than the DE. But it plays an important role in the PS and cannot be neglected.[1] In a two-dimensional $N \times N$ lattice, each site has a single t_{2g} spin, and only a fraction $1-x$ of the sites are each occupied by an e_g spin, corresponding to, e.g., $T_{1-x}D_xMnO_3$, where T is a trivalent rare earth and D is a divalent alkaline.

We use a simplified method based on the DE in real space, instead of diagonalizing the Hamiltonian matrix to get the eigenvalues and energy band information. We suppose $J_H = \infty$, and the e_g spin is always parallel to the t_{2g} spin of the same site. At each step, a site is selected at random. Then the dynamic procedure consists of two consecutive steps: First is the spin flip with a certain probability. We count the number of possible double exchanges n. If the site contains one e_g electron, then n equals the number of its NN sites with a parallel spin t_{2g} core but without an e_g electron. Otherwise, n equals the number of its NN sites containing a parallel e_g spin. After n is obtained, we accordingly give the energy contribution $E_1 = -nt$. Thus, whether $x \to 0$ or 1, the kinetic energy of the e_g electron vanishes. We also calculate the energy contribution E_2 of the AF superexchange and the external field. Then, the energy is recalculated after the t_{2g} spin (and the e_g spin, if there is one) is (are) flipped. If the energy is lowered, the spin(s) is (are) flipped. Otherwise the probability of flip is given by

$$W = \exp\left(-\frac{1}{k_B T}\Delta E\right), \tag{2}$$

where ΔE is the energy difference. The second step is the e_g electron hopping: Whether the flip in the first step is successful or not, one of the possible hopping processes that is considered in the energy calculation is randomly chosen and realized. Here the fermion property and the Pauli exclusion rule are considered. Then in the next time step, another site is chosen at random and the above-mentioned procedure is repeated. A single MC step consists of $N \times N$ such unit procedures.

Rather than the strict eigenstates, we consider the tran-

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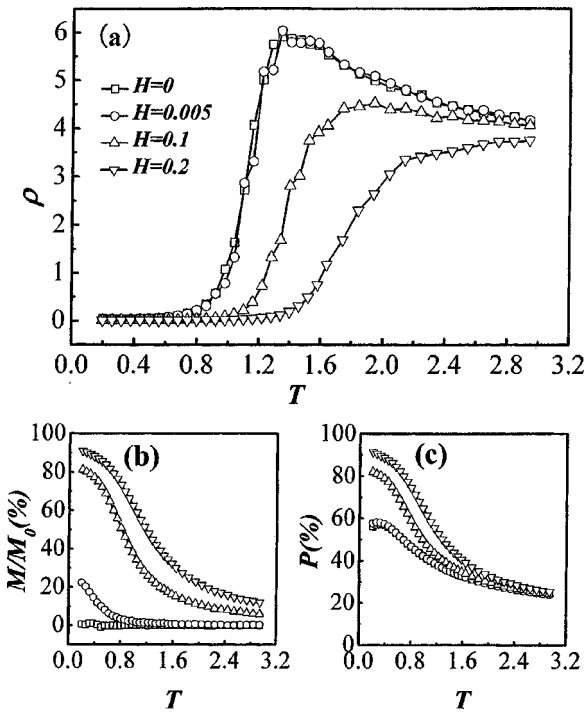

FIG. 1. (a) The system resistivity, (b) the system magnetization M (divided by the saturated magnetization M_0), and (c) the fraction of metallic resistors are shown as a function of temperature for doping percentage $x=0.5$ and external field $H=0$ (squares), 0.005 (circles), 0.1 (triangles), and 0.2 (inverted triangles).

sitions among simpler spin configurations. The METROPOLIS update is realized by e_g spin flip and electron hopping, of which the probabilities are determined by the Hamiltonian. It is clear that the physical essence of DE and the other interactions is represented in our model. The motivation is as follows: Because in experiments the length scale of percolation in the phase-separated manganite system usually far exceeds the lattice constant and even reaches μm scale, the lattice scale in theoretical work needs to be enlarged. Much larger systems can be studied microscopically by our model, and the intrinsic physics is kept and presented in a clear way. As will be shown in the following, the model presents MIT and CMR, which are in agreement with experiments qualitatively.

Beginning from an initial spin configuration, the system is believed to be in equilibrium after a relaxation process (in our study of a 100×100 system, usually 300 MC steps are enough). Then a resistor network is constructed based on the spin configuration in the following way. The t_{2g} electrons are divided into two groups: those with three or more of their nearest neighbors having the same spin direction are in group 1 and the remaining ones are in group 2. Then if a pair of NN spins are both in group 1, a metallic resistor is established between them; otherwise an insulating resistor is established instead. The total resistivity is calculated using the method of Ref. 10. The results are obtained by taking the average of many independent runs. Here some arbitrariness remains in the choice of the resistivity functions. In the present work for the metallic resistors $\rho_M(T)=0.005+0.0025T^2$, and for the insulating resistors $\rho_I(T)=0.5Te^{6/T}$. The effect of the specific choice of $\rho_M(T)$ or $\rho_I(T)$ will be discussed in the following. Because the metallic (insulating) resistors aggregate into the metallic (insulating) clusters much larger than the lattice constant, in fact here are course-grained resistor networks and the Ohmic rule is used as a simplified method, although real nanometer-scale transport should be calculated within a quantum treatment.[11] Different from a randomly generated network, the present network obtained from the DE can present the cooperative nature of the manganite system, which leads to a different type of percolation, as shown in the following.

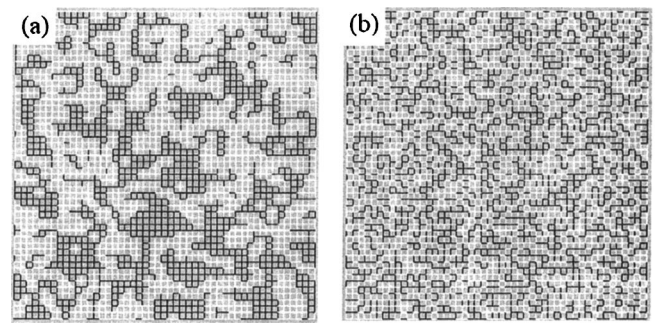

FIG. 2. (a) A 50×50 part of a resistor network generated from a 100×100 system with $x=0.5$, $T=1$, and $H=0$, in comparison with (b) the picture of standard percolation. Both of them have the fraction of metallic resistors $p=0.41$. Black bonds correspond to metallic resistors, and white bonds correspond to insulating resistors.

For $x=0$ or 1, the system has purely AF interaction, with the resistivity simply described by $\rho_I(T)$. For $x=0.5$, Fig. 1(a) shows the resistivity as a function of temperature in different external field H. With $H=0$, a MIT is observed: For high temperature, it is a paramagnetic insulating system, with the resistivity dominated by $\rho_I(T)$. As the temperature decreases, ferromagnetic (FM) regions begin to emerge, and growing domains are visible. However, the directions of the polarization of the different FM domains are random in a zero field, giving a zero total magnetization for any temperature. The magnetic transition is revealed when an external field is applied [Fig. 1(b)], as studied in the following. At very low temperature (relative to the MIT point) and with zero field, in this inhomogeneous dynamic system weak AF order can temporarily emerge in the regions where either there are less e_g electrons, or the e_g electrons aggregate and have less mobility. However, the percolation of metallic resistors still holds in $x=0.5$ situation. (In our simulation, the fraction of

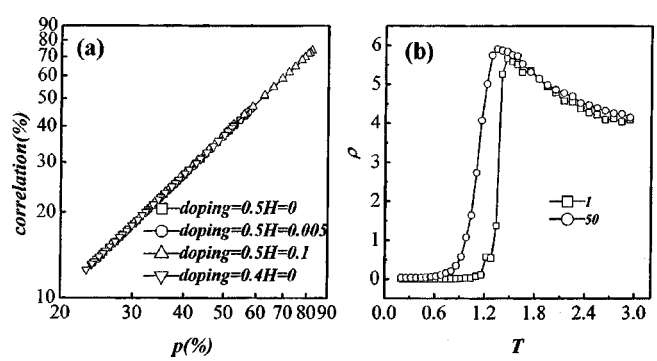

FIG. 3. (a) The $\log_{10}-\log_{10}$ graph of the correlation of resistors as a function of metallic resistor fraction p, with $x=0.5$, $H=0$ (squares); $x=0.5$, $H=0.005$ (circles); $x=0.5$, $H=0.1$ (triangles); $x=0.4$, $H=0$ (inverted, triangles). The solid line with slope 1.4 serves as the guide to the eye. (b) The system resistivity is plotted as a function of temperature. Circles represent $\rho_M^{(1)}(T)=0.005+0.0025T^2$ and $\rho_I^{(1)}(T)=Te^{3/T}$, while squares represent $\rho_M^{(2)}(T)=\rho_M^{(1)}(T)/50$ and $\rho_I^{(2)}(T)=\rho_I^{(1)}(T)$.

metallic resistors $p=0.51$ for $x=0.5$, $T=0$, and $H=0$, larger than the threshold $p_c \approx 0.36$.)

As shown in Fig. 1(c), p can be obtained microscopically, as a function of T, x, and H. Generally speaking, p is larger when x approaches 0.5, where the one-orbital double exchange has the maximal effect. When a very small field (in Fig. 1, $H=0.005$) is applied, the magnetization M is significantly enhanced at low temperature [Fig. 1(b)] and this reveals the magnetic transition. However, the resistivity as a function of temperature is largely unaffected [Fig. 1(a)], except for the region of very low temperature. Actually, this is because p remains largely unchanged, as shown in Fig. 1(c). The deeper reason is that a small field can make parallel the directions of the already formed FM domains, but it is not sufficient to form new FM domains.[12] In the present model with $x=0.5$, another effect of the weak field is the destruction of weak AF order at very low temperature, and the curves of $p(T)$ and $\rho(T)$ are influenced accordingly. When a larger field is applied, both M and p will be higher [Figs. 1(b) and 1(c)], and the magnetoresistance effect can be observed, with the MIT point shifting to higher temperature.

To better understand the relationship between the percolation and the MIT, we move on to discuss the second issue: the nature of the percolation. (1) In Fig. 2, a snapshot of the resistor network obtained from the system of $x=0.5$, $T=0.5$, and $H=0$ is given, in comparison with a SP picture of the same value of p. Black bonds represent metallic resistors and white bonds represent insulating resistors. The difference is obvious: In Fig. 2(a), there are many easily identifiable compact clusters of purely metallic resistors or insulating resistors. This directly leads to (2) a percolation threshold 0.36, smaller than the threshold 0.5 of the standard percolation. (3) The correlation of NN resistors is also studied. Each resistor has six nearest neighbors, and we give value 1 to metallic resistors and value 0 to insulating resistors. In the SP situation, the average NN correlation $C=p^2$. In the present percolation system, we observe a stronger correlation $C' \approx p^{1.4}$, as shown in Fig. 3(a). From the data collapse we have gathered, we find that this is a universal relationship. It suggests that the structures of different systems with varying x, T, and H are unified into a single class of percolation, with a unique percolation threshold, $p_c=0.36$.

Now we need to clarify, since the structures are unified, why the resistivity as a function of T may look so different and why the fraction p corresponding to the MIT point may vary, though slightly. A very curious experimental example is that, in the system of $Sm_{0.55}Sr_{0.45}MnO_3$,[13] the MIT appears to be first order in a zero field, while it becomes much less abrupt in a 4 kOe field. It can be explained in the present model by taking into consideration the fact that the resistivity also depends on $\rho_M(T)$ and $\rho_I(T)$. We illustrate this effect in two examples: As shown in Fig. 1(a), the MIT corresponding to $x=0.5$ and $H=0.1$ is much less abrupt than the curve in zero field, and the MIT point shifts to $p \approx 0.31$, which means that the system is already metallic below the percolation threshold. The first difference is mainly because $\rho_I(T)/\rho_M(T)$ is much smaller in the $H=0.1$ curve near the MIT, and the shifted MIT point can be attributed to the reduced ratio of

$$\left| \frac{d\rho_I(T)}{dT} \Big/ \frac{d\rho_M(T)}{dT} \right|.$$

By contrast, very abrupt MIT can also be achieved: With $x=0.5$ and $H=0$, if at the MIT point, $\rho_I|_{\mathrm{MIT}}$ is chosen to be larger than $\rho_M|_{\mathrm{MIT}}$ by four magnitudes, as shown in Fig. 3(b), the MIT is continuous and less abrupt. However, if $\rho_M(T)$ is reduced to $1/50$ of the previous function (this is approximately what is used by Mayer *et al.* in Ref. 4, according to the experiment of Ref. 12), as shown in Fig. 3(b), the MIT becomes much more abrupt.

To summarize, we report a METROPOLIS MC study of the one-orbital double exchange lattice model. A 100×100 system is used to display microscopically the phase competition and percolation. Given an initialization of spin configuration, temperature T, and external field H, a resistor network is constructed based on the spin configuration after a relaxation process. Much attention is paid to the phase competition and percolation. The MIT is described by the competition between a fraction p of metallic resistors and a fraction 1-p of insulating resistors. Our results can be summarized in three aspects: (1) p can be obtained as a function of T and H, with different doping percentage x. (2) The microscopic details of the percolation are presented from system evolution. Based on the microscopic simulation of the percolation, systems with different x, T, and H are unified to a single class of percolation, with the structure only determined by p. (3) The system resistivity is then given by p and the values of the metallic and insulating resistivity.

The authors thank the Natural Science Foundation of China (50332020, 10021001) and National Key Projects for Basic Research of China (2002CB613303, 2004CB619004).

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