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Long-range hopping correlation and colossal magnetoresistance in doped manganites

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Abstract. The nature of the paramagnetic-insulator-to-ferromagnetic-metal transition and the associated colossal magnetoresistance (CMR) in doped manganites is re-investigated on the basis of the double-exchange model by using a Monte Carlo technique. We calculate the temperature dependence of the magnetic susceptibility and resistivity by taking account of the magnetic fluctuation together with the long-range hopping correlation. The results show Curie–Weiss behaviour of the susceptibility, consistent with a sharp peak in the resistivity near the Curie temperature T_c. The estimated value of T_c and the magnitude of the CMR in the magnetic field are in good agreement with experiments. Our results clearly demonstrate the importance of the magnetic fluctuation and the long-range hopping correlation in the CMR manganites.

1. Introduction

The recent observation of colossal magnetoresistance (CMR) in mixed-valence manganites has led to there being a great deal of interest in achieving an understanding of their unusual magnetotransport properties [1]. One of the most prominent features of these materials is a close relationship between the metal–insulator (MI) and ferromagnetic-to-paramagnetic transitions. The generic behaviour is well understood within the framework of the double-exchange (DE) theory [2], where the spin–spin correlation between the localized spins is crucial to determining the amplitude for hopping of the conduction electrons between neighbours and, in particular, the resistivity (ρ) near the Curie temperature, T_c [3]. In previous studies, however, even though it was considered to be responsible for the insulating behaviour, i.e., $\mathrm{d}\rho/\mathrm{d}T < 0$, in the paramagnetic state, the spatial spin correlation was not treated properly, so the MI transition and the associated CMR near T_c could not be understood within mean-field-type approaches [4, 5].

Recently Millis *et al* [6] argued that DE model alone is not enough to explain a sharp change in ρ near T_c and proposed that the lattice polaron formation arising from the dynamic Jahn–Teller distortion is essential for the MI transition and CMR. Despite the efforts made in [7, 8], the lattice polaron view has not been successful in explaining the enormous drop of magnetoresistance. Although the Jahn–Teller distortion is quite important for the understanding of overall trends in the manganite physics, there has been an increasing realization [9–12] that lattice polaron formation is not sufficient to explain the transport

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properties in connection with the CMR phenomena. On the other hand, there are numerous reports [14–17] on the importance of the magnetic polaron formation caused by the spin-disorder scattering inherent to the DE model. According to this scattering mechanism, the charge carriers moving in the slowly fluctuating spin background form magnetic polarons around T_c due to the robust fluctuation in the spin correlation, $\langle \vec{S}_i \cdot \vec{S}_j \rangle$. Several authors [3–5] also suggested a possible crucial role of the spin fluctuations in the resistivity and the CMR phenomena near and above T_c. However, in these works [3–5, 14–17] the importance of the magnetic fluctuations in the long-range hopping correlations has not been fully appreciated.

In this paper, we report the results of our studies on the temperature dependence of the resistivity, taking account of the magnetic fluctuations together with the long-range hopping correlations. In order to include the effects of the fluctuations of the localized t_{2g} spins of Mn atoms, we adopt an unbiased Monte Carlo method [17–19] on a three-dimensional (3D) DE system. From the results, it is shown that the resistivity from the long-range hopping calculation has a sharp peak near T_c and decreases exponentially above T_c with increasing temperature in the paramagnetic phase. The magnitudes of both the magnetoresistance ratio and the estimated T_c are in good agreement with experimental observations. This is the first result demonstrating the importance of magnetic fluctuations and the long-range hopping correlations in CMR manganites.

2. Model and calculations

The following simplified model Hamiltonian for understanding the doped manganites is a single-orbital DE Hamiltonian with a magnetic field [2, 6]:

$$\mathcal{H} = -\sum_{\langle ij \rangle} (t_{ij} c_i^+ c_j + \text{h.c.}) - h \sum_i S_i^z - \mu \sum_i c_i^+ c_i \tag{1}$$

where the operator c_i^+ creates a spinless conduction electron at site $\vec{R}_i$, $\vec{S}_i$ refers to the localized t_{2g} spins, h is an external magnetic field, and μ is the chemical potential. In the limit of strong Hund's coupling the hopping amplitude is given by

$$t_{ij} = t\left(\cos\frac{\theta_i}{2}\cos\frac{\theta_j}{2} + \sin\frac{\theta_i}{2}\sin\frac{\theta_j}{2}\mathrm{e}^{\mathrm{i}(\phi_i - \phi_j)}\right) \tag{2}$$

with the polar angles $\{\theta_i, \phi_i\}$ characterizing the orientation of the localized spin:

$$\vec{S}_i = S(\sin\theta_i \cos\phi_i\, \hat{x} + \sin\theta_i \sin\phi_i\, \hat{y} + \cos\theta_i\, \hat{z}).$$

For simplicity, we assume that the azimuthal angle ϕ_i rotates independently [6]. Thus, it is possible to rewrite t_{ij} in the familiar form $t_{ij} = t\cos(\theta_{ij}/2)$. The calculation of the partition function of the present model is based on a finite-temperature Monte Carlo technique introduced by Yunoki *et al* [18]. Indeed, this method has been successfully applied in the study of the phase separation [18, 19] and the magnetic phase diagram of the electronic model for manganites [17]. Thermodynamic quantities of interest are obtained directly from the ensemble average of the spin configurations and the eigenvalues of the Hamiltonian. The actual calculations were performed for $L = 6$ and $L = 4$ cubic lattices with periodic boundary conditions. The carrier density $\langle n \rangle \approx 0.5$, i.e., the hole density $x = 1 - \langle n \rangle$; this was obtained by fixing $\mu = 0.0$. Unless stated otherwise, we take $t = 1$, $|S| = 1$, and $L = 6$.

3. Monte Carlo results

3.1. Ferromagnetic-to-paramagnetic transition near T_c

Figure 1 shows the temperature dependence of the magnetization:

$$M \equiv \left\langle \sum_i S_i^z \right\rangle \Big/ L^3$$

denoted by solid circles and the corresponding magnetic susceptibility:

$$\chi \equiv (\langle M^2 \rangle - \langle M \rangle^2)/k_B T$$

represented by open squares. As clearly seen in figure 1, the calculated χ shows a steep peak near T_c in accord with the onset of the ferromagnetic transition. The dashed line is a mean-field prediction of $\chi \sim \langle S^2 \rangle/(T - T_c)$ in terms of the magnetic polaron formation [14, 15] caused by the spin-disorder scattering. The Curie–Weiss behaviour of χ allows us to extrapolate the mean-field value of T_c: it is $0.13t$, which is consistent with the results from the high-temperature expansions [13] as well as other theoretical methods [14–18]. If we take values of the bandwidth between 0.1 and 0.3 eV from the literature [4, 18], our estimate for T_c lies between 150 and 450 K, which coincides with the range observed experimentally. In contrast, the previous estimate [6] of $T_c \approx 0.3$ eV ≈ 3600 K was much higher due to the neglect of the thermal fluctuation effect [4] as well as the large value of the exchange spin coupling $J \sim 0.1$ eV used for the 3D Heisenberg ferromagnet model. Indeed, recent study [20] of the spin-wave dispersion for $La_{0.7}Pb_{0.3}MnO_3$ ($T_c = 355$ K) evaluated $J \sim 0.002$ eV and $T_c \approx 410$ K. Therefore, it is evident that the DE model alone can account for the value of T_c, the Curie–Weiss behaviour of χ, and a sharp drop of the magnetization near T_c.

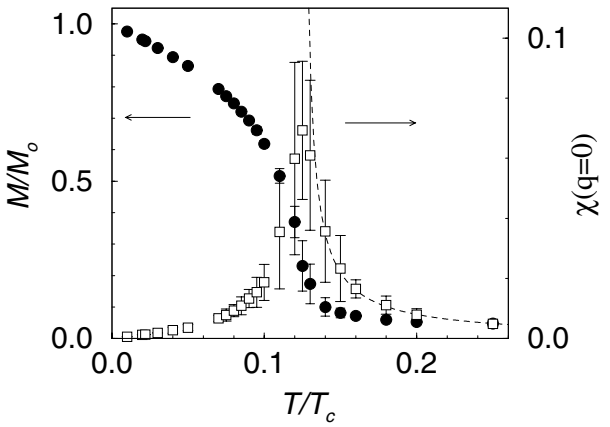

Figure 1. Magnetization M and magnetic susceptibility $\chi \equiv (\langle M^2 \rangle - \langle M \rangle^2)/T$ as functions of temperature. The dashed line is a mean-field prediction for χ of the form $\langle S^2 \rangle/(T - T_c)$ from reference [13].

Figure 2 shows the temperature dependence of the average hopping integral $\langle t_{ij} \rangle = \langle \cos(\theta_{ij}/2) \rangle$ for the Monte Carlo simulation and the mean-field prediction [3] in the limit of $S = \infty$. We see that the Monte Carlo data for $\langle t_{ij} \rangle$ are a smooth and continuous function of temperature for the whole range of T. Note, however, that $\langle t_{ij} \rangle$ varies even in the spin-disorder regime and, in particular, the curvature, i.e., $\partial^2 \langle t_{ij} \rangle / \partial T^2$, changes sign near T_c which is at variance with the mean-field prediction. This result implies that a hopping fluctuation has a maximum as the curvature approaches zero in the vicinity of T_c since it is related to the second

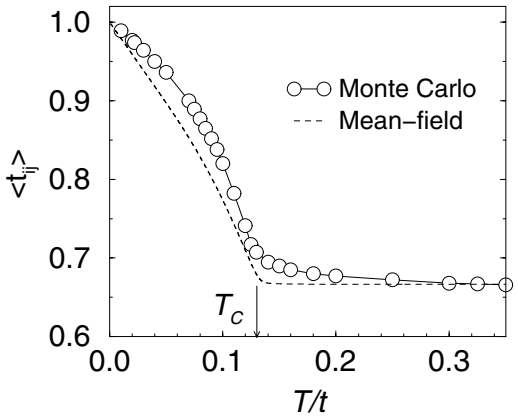

Figure 2. The temperature dependence of the hopping integral $\langle t_{ij}\rangle = \langle\cos(\theta_{ij}/2)\rangle$ for the Monte Carlo and the mean-field predictions. T_c is indicated by a downwards-pointing arrow.

derivative of M via the DE mechanism, leading to a sharp peak in $\chi(T_c)$ as shown in figure 1. In other words, the carriers are trapped by spin-disorder scattering in the spatially fluctuating hopping potential due to local deviations of the ferromagnetic surroundings, thereby producing a localization in the paramagnetic phase [11, 14]. In the high-temperature limit of $T \sim 3T_c$, we obtain the $\langle t_{ij}\rangle = 2/3$, leading to the reduction of the effective bandwidth in the density of states [26].

3.2. Resistivity in the absence of hopping correlation

The resistivity can be determined by using the phenomenological theory of the Drude formula $\rho = m^*/xe^2\tau$, where x is the concentration of hole carriers, m^* is the effective mass of a carrier, and τ is the relaxation time. In the DE model, $1/m^*$ is set by the effective bandwidth proportional to $\langle t_{ij}\rangle$. Kubo and Ohata [3] adopted an approximation for the imaginary part of the electron self-energy Σ, in the presence of the spatially fluctuating random potential U caused by the spin-disorder scattering:

$$\frac{1}{\tau} = \mathrm{Im}\,\Sigma \approx \frac{1}{2m^*k_BT}\left\langle\left(\frac{\partial U}{\partial t_{ij}}\right)^2\right\rangle\tau_c \tag{3}$$

where τ_c is the characteristic time in which the coherent random force persists. The random potential of mainly magnetic origin is closely related to the fluctuation of the hopping integral due to the DE mechanism induced by the spin-disorder scattering. For instance, in the presence of strong Hund's rule coupling J_H, the random potential arising from the spin-flip scattering of the form $-J_H\vec{S}\cdot\vec{\sigma}$ between localized Mn t_{2g} spins and the spins of the e_g electrons [11] can be written as $U \sim J_HS(1-\cos(\theta_{ij}))$, where θ_{ij} is the angle between the spins of the two Mn ions.

According to the phenomenological study of Kubo and Ohata [3], $\langle(\partial U/\partial t_{ij})^2\rangle$ can probably be replaced by $\langle(\Delta t_{ij}\,k_F)^2\rangle$, where Δt_{ij} is the fluctuation of the hopping integral defined as $\Delta t_{ij} \equiv t[\cos(\theta_{ij}/2) - \langle\cos(\theta_{ij}/2)\rangle]$. In our problems, k_BT may be replaced by E_F and τ_c by am^*/k_F, where a is a lattice constant and k_F is the Fermi wave vector. The resulting resistivity is estimated as

$$\rho \sim \frac{a}{xe^2k_F}\frac{\langle(t_{ij})^2\rangle - \langle t_{ij}\rangle^2}{\langle t_{ij}\rangle^2}. \tag{4}$$

Within this phenomenological treatment, Kubo and Ohata showed that the resistivity behaves as $\rho \sim 1 - M^2$ within the framework of the molecular-field approximation, which is consistent with the recent dynamic mean-field theory given by Furukawa [4]. These results qualitatively help us to understand the ferromagnetic metallic state of the DE model via a spin-disorder scattering mechanism.

To study the hopping-fluctuation effects in the vicinity of T_c, we plot the T-dependence of the resistivity as a function of T/T_c for the Monte Carlo and the mean-field calculations in figure 3. We see that the resistivity from the Monte Carlo calculation shows a similar behaviour to that from the mean-field calculation below and above T_c. A small peak structure of the Monte Carlo calculation of ρ at temperature very close to T_c must come from the fluctuation effects of the background localized spins which are ignored in the mean-field calculation. However, the present results fail to reproduce the experimental observations of the insulating behaviour with $\mathrm{d}\rho/\mathrm{d}T < 0$ in the paramagnetic state. One reason for the discrepancy between this phenomenological theory and experiments is that the effects of the shortest hopping correlation and its fluctuation are not sufficient to trap the charge carriers in the conduction bands above T_c.

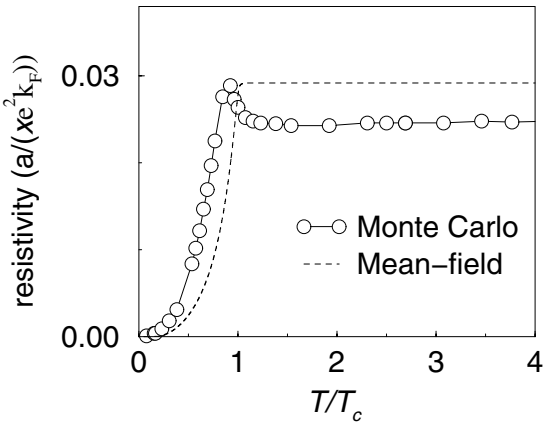

Figure 3. The T-dependence of the resistivity in the absence of the hopping correlation from equation (4) for mean-field and Monte Carlo calculations.

3.3. Long-range hopping correlation and resistivity

Since a finite-range ferromagnetic spin correlation persists even above T_c, however, the contribution from long-range hopping correlations to the spin-disorder scattering which is responsible for the carrier localization cannot be entirely ignored in our problems. Thus, we may conjecture that the inverse relaxation time can be written as

$$\frac{1}{\tau} \approx \frac{a}{k_F \langle t_{ij} \rangle} \left\langle \sum_{\delta_1, \delta_2, i, R} \Delta t_{i,i+\delta_1} \, \Delta t_{i+R,i+R+\delta_2} \right\rangle = \frac{a}{k_F \langle t_{ij} \rangle} (\Gamma - \langle t_{ij} \rangle^2) \tag{5}$$

where Γ is the long-range hopping correlation defined as

$$\Gamma \equiv \frac{1}{(3L^3)^2} \left\langle \sum_{\delta_1, \delta_2} \sum_{i, R} t_{i,i+\delta_1} t_{i+R,i+R+\delta_2} \right\rangle. \tag{6}$$

Here R represents sites on the 3D lattice, and δ_1 and δ_2 are the unit vectors connecting its nearest-neighbour sites. In the presence of the long-range hopping correlation, the corresponding

resistivity is written as

$$\rho \approx \frac{a}{xe^2k_F}\frac{\Gamma - \langle t_{ij}\rangle^2}{\langle t_{ij}\rangle^2}. \tag{7}$$

It is worth noting that this formula is nothing but the generalization of equation (4), implying that the fluctuation of the hopping correlation defined as $\Delta\Gamma \equiv \Gamma - \langle t_{ij}\rangle^2$ localizes the charge carriers moving in a fluctuating spin background in the paramagnetic state.

Figure 4(a) shows the T-dependence of Γ and $\langle t_{ij}\rangle^2$ near and above T_c. Since the spatial hopping correlation is greater than the $\langle t_{ij}\rangle^2$, this allows us to measure the fluctuation of the hopping correlation $\Delta\Gamma$. The temperature dependence of $\Delta\Gamma$ is shown in figure 4(b). The fluctuation reaches a maximum very close to T_c and is reduced more rapidly below and above T_c. This behaviour is closely related to the peak structure of the relaxation time and the resistivity.

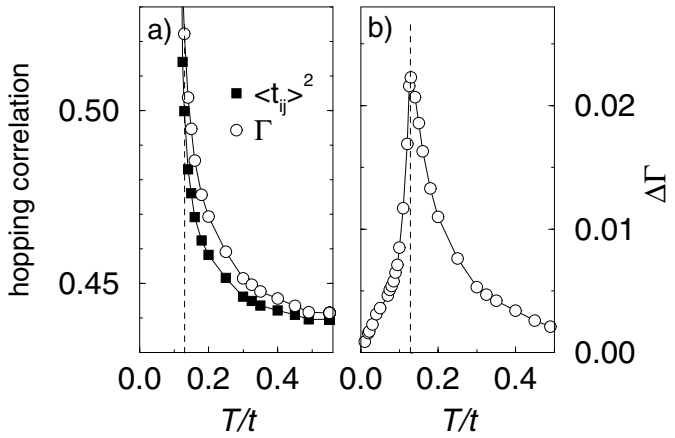

Figure 4. (a) The long-range hopping correlation Γ and $\langle t_{ij}\rangle^2$ in the vicinity of T_c (see the text). (b) The temperature dependence of the long-range hopping correlation defined as $\Delta\Gamma \equiv \Gamma - \langle t_{ij}\rangle^2$. The dashed lines indicate T_c.

The T-dependence of the corresponding ρ and τ^{-1} in the long-range hopping regime is shown in figure 5. With increasing temperature, the curves for ρ and τ^{-1} each increase to a maximum near T_c (marked as a dashed line) and then decrease above T_c. The rapid decrease of ρ below T_c and the insulating behaviour ($\mathrm{d}\rho/\mathrm{d}T < 0$) above T_c are in good agreement with experiments. Similar behaviours are obtained from the simulation with different doping levels. Moreover, the resistivity shown in the inset of figure 5 follows the Mott variable-range hopping law $\rho = \rho_0 \exp((T_0/T)^{1/4})$ in the semiconducting region [21]. A detailed study of the resistivity above T_c made by putting together experimental measurements for $La_{1-x}Ca_xMnO_3$ single crystals and the present Monte Carlo results will be published elsewhere [22]. From the slope of the fitting curve, we estimate the localization length, which is the size of the large bound magnetic polaron, to be 8 Å from $k_BT_0 = 24/(\pi N(E_F)\xi^3)$ [11, 14, 21]. This value of ξ is physically plausible, since it exceeds the average Mn^{3+}–Mn^{4+} spacing of about 4 Å. Thus it is argued that a simple physical picture in accord with our Monte Carlo studies is that of carrier localization due to the fluctuation of the long-range hopping correlation induced by fluctuating spin backgrounds. However, more precise determination for ξ requires a finite-size scaling analysis, which is beyond the scope of this work due to the computational limitations. Indeed, there is some finite-size effect in the calculated M above T_c [23].

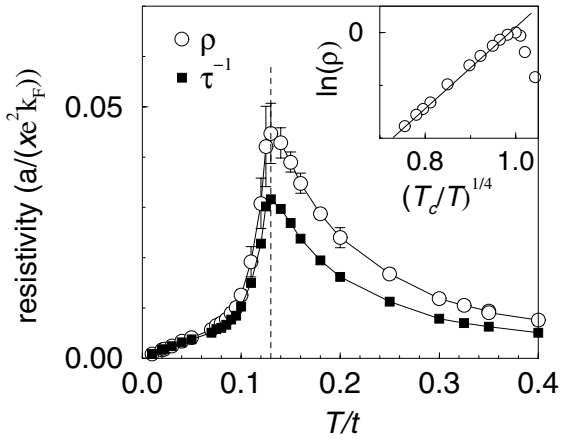

Figure 5. The T-dependence of the resistivity and the relaxation time in the long-range hopping regime. The dashed lines indicate T_c. In the inset, we show $\ln(\rho)$ versus $(T_c/T)^{1/4}$.

3.4. Colossal magnetoresistance

For completeness, we study the effect of the magnetic field on the MI transition and CMR phenomena. The temperature dependence of the resistivity, i.e., $\rho \approx \Delta\Gamma/\langle t_{ij}\rangle^2$, for zero magnetic field and $h = 0.02$ for $L = 6$ are shown in figure 6. Note that the estimate of the magnetic field $h = 0.01t$ corresponds to 5 T if we take $t = 0.2$ eV for $T_c \approx 300$ K. The applied magnetic field greatly reduces the resistivity peak around T_c and drives its transition to a higher temperature due to the suppression of the spin-disorder scattering. The magnetoresistance obtained, defined as $\Delta\rho/\rho \equiv (\rho(0) - \rho(h))/\rho(0)$, at $T_c = 0.13t$ is about 70% which is in good agreement with the experimental observation.

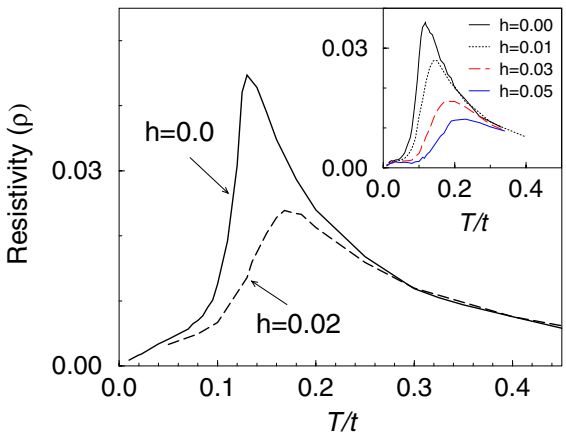

Figure 6. ρ versus T for several values of the magnetic fields and $L = 4$. In the inset, we show the magnetoresistance ratio, $\Delta\rho/\rho$, as a function of T for various values of the magnetic field.

Similar results are also obtained for smaller systems with $L = 4$. We plot the T-dependence of the resistivity for different values of the magnetic field in the inset of figure 6. The maximum magnetoresistance obtained here, $\Delta\rho/\rho \approx 54\%$, 86%, and 94% for $h = 0.01$, 0.03, and 0.05, respectively, compares well with most experimental measurements.

4. Discussion

It is interesting to compare our Monte Carlo results with recent calculations of Ishizaka and Ishihara [5] and Millis *et al* [6] since they used an expression similar to equation (7) except that they derived a four-spin correlation instead of $\Delta\Gamma$. The four-spin correlation obtained from the lowest-order perturbational treatment within the memory function method [24, 25] is written as

$$\Delta C \approx \sum_{\langle ij\rangle,R} \langle \vec{S}_i \cdot \vec{S}_j \vec{S}_{i+R} \cdot \vec{S}_{j+R} \rangle B(R) \tag{8}$$

where $B(R)$ is the decaying oscillatory function. To have a better understanding of which spatial correlation function is responsible for the observed resistivity behaviour, we reproduce ΔC obtained through Monte Carlo calculations and compare the result with $\Delta\Gamma$ shown in figure 4. The temperature dependence of the correlation functions normalized by the values at the peak position, $\Delta C/\Delta C_{peak}$ and $\Delta\Gamma/\Delta\Gamma_{peak}$, are plotted in figure 7. ΔC shows an anomalous peak far below T_c while $\Delta\Gamma$ shows a clear peak near T_c. A similar upturn behaviour in ΔC has been reproduced by different methods, e.g., the Schwinger boson approach [5] and the spherical model approximation [6].

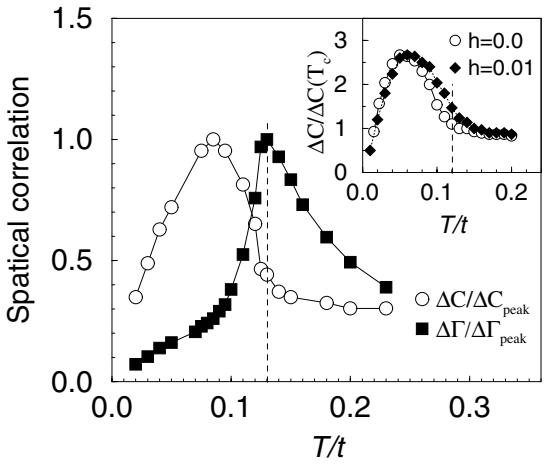

Figure 7. The spatial four-spin and long-range hopping correlations normalized by the values at the peak position, $\Delta C/\Delta C_{peak}$ and $\Delta\Gamma/\Delta\Gamma_{peak}$, respectively. The inset shows the normalized $\Delta C/\Delta C(T_c)$ for zero magnetic field and for a field of $h = 0.01$ for $L = 4$. The dashed lines indicate T_c.

It is well known that the correlation function responsible for the transport property is greatly reduced under a magnetic field since it suppresses the spin-disorder scattering of the carriers. To investigate the influence of the magnetic field on the four-spin correlation function, we plot the normalized $\Delta C/\Delta C(T_c)$ as a function of T for zero magnetic field and $h = 0.01$ for $L = 4$ in the inset of figure 7. However, ΔC under the magnetic field does not show a large reduction in the robust ferromagnetic state, even for $T \ll T_c$, or a shift of the peak position to higher temperature. In other words, the four-spin correlation is not directly related to the resistivity of the DE model via a spin-disorder scattering mechanism, or more precisely magnetic polaron formation.

In fact, on the basis of the result for ΔC, Millis *et al* [6] showed that the resistivity is still increased below T_c with decreasing T and, taking this together with the overestimated

mean-field T_c as discussed above, they suggested that an additional physical mechanism, such as lattice polaron formation, is necessary to reproduce the observed resistivity and CMR phenomena. Unlike the previous results of references [5] and [6], however, our calculations of $\Delta\Gamma$, $1/\tau$, and ρ are remarkably consistent with the experimental observations. The discrepancy between the present and the earlier works is attributable to the unusual upturn behaviour in ΔC shown in figure 7, which may be an artifact of the perturbational treatment or memory function method, because the spin correlation, $\langle \vec{S}_i \cdot \vec{S}_j \rangle$, shows no anomaly in the temperature region of interest [15, 18].

It is also worth noting that Calderón *et al* [26] calculated the conductance by utilizing a different Monte Carlo method with the Kubo formula. They found an absence of insulating behaviour above T_c for $x > 0.1$, and that the system is metallic for all temperatures. This result is similar to the previous mean-field results obtained by Furukawa [4] and Kubo and Ohata [3] as well as our Monte Carlo results for the shortest hopping correlation of scattering, i.e., $\vec{R}_i = 0$. However, the physical mechanism leading to the T-dependence of the resistivity in the paramagnetic state is completely different from our result. The driving source of the localization in our Monte Carlo calculation is the combined effect of the hopping fluctuation and its long-range correlation. Indeed, some authors [3, 5, 25] point out that the contribution from $\Delta\Gamma$ for $\vec{R}_i \neq 0$ cannot be neglected in the vicinity of T_c and may be responsible for a decrease in resistivity above T_c. On the other hand, the local spin-fluctuation effect is fully taken into account in reference [23]; however, the fluctuation effects of the long-range hopping correlation are completely ignored in their results. Without any additional degrees of freedom such as electron–lattice coupling, our numerical studies clearly demonstrate that a spin-disorder scattering induced by a long-range hopping correlation and its fluctuation is a possible origin of the metal–insulator transition and the associated CMR phenomena in doped manganites.

5. Summary

In summary, we investigated the driving mechanism of the MI transition within the DE model by taking account of the spin fluctuation and the long-range hopping correlation using a Monte Carlo technique. The calculated T-dependence of the resistivity shows a sharp peak near T_c as well as an insulating behaviour above T_c, which are consistent with experimental observations. The estimated T_c-value and the magnitude of the CMR in the magnetic field are in good agreement with experimental results. We suggest that the long-range hopping correlation is important in understanding the transport properties of the CMR manganites, and furthermore claim that the CMR phenomenon is probably related to the magnetic polaron formation driven by the spin-disorder scattering. We hope that our results stimulate a wider discussion and more experimental investigation in this direction.

Acknowledgments

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