Simulation of Temperature-Dependent Resistivity for Manganite $La_{1/3}Ca_{2/3}MnO_3$*

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The resistivity of the heavy-doped $La_{1/3}Ca_{2/3}MnO_3$ (LCMO) is simulated using a random resistor network model, based on a phase separation scenario. The simulated results agree well with the reported experimental data, showing a transition from a charge-disordered (CDO) state embedded with a few ferromagnetic (FM) metallic clusters to a charge-ordered (CO) state, corresponding to the transition from a high-temperature paramagnetic (PM) insulating state to a low-temperature antiferromagnetic (AF) insulating state. Furthermore, we find that the number of AF/CO clusters increases with decreasing temperature, and the clusters start to connect to each other around 250 K, which causes percolating in the system. The results further verify that phase separation plays a crucial role in the electrical conductivity of LCMO.

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Manganite doped $LaMnO_3$ has been intensively studied due to its rich phase diagram and the colossal magnetoresistance effect.[1−8] For $La_{1-x}Ca_xMnO_3$, at $x = 0$, parent compound $LaMnO_3$ is insulating at all temperatures. It is paramagnetic at high temperature, but at low temperature it becomes antiferromagnetic. At $\sim 0.2 < x < 0.5$, the compound undergoes a phase transition from the low-temperature ferromagnetic (FM) phase to the high-temperature paramagnetic (PM) phase with a metal-to-insulator transition (MIT) of resistivity. Furthermore, the material generally exhibits a paramagnetic (PM) charge-disordered (CDO) state at high temperature and an antiferromagnetic (AF) charge-ordered (CO) state at low temperature for $\sim 4/8 < x < 7/8$.[9,10] Many publications have given analyses of MIT behaviour of the transport properties of the slight-doped $La_{1-x}Ca_xMnO_3$ such as $\sim 0.2 < x < 0.5$. Mayr *et al.*[11] obtained the good qualitative results of MIT behaviour by using a random resistor network based on the mixed-phase theory. In our previous work, we presented a calculation method based on the random network model by introducing a breadth-first traversal (BFT) algorithm to simulate the MIT behaviour, and good quantitative agreements with experimental data have been obtained.[12,13]

However, few quantitative studies have been carried out on the resistivity of $x = 2/3$ heavy-doped compound since the transport properties of CO state remain unsolved and theoretical calculation of the resistivity is notoriously difficult. Hence, it is necessary to understand the true physical mechanism of CO state formation of the manganite which undergoes a PM charge-disordered state to an AF charge-ordered state transition. Zhou *et al.*[14] have measured electron spin resonance spectra of manganese perovskite $La_{1/3}Ca_{2/3}MnO_3$ (LCMO) and observed the existence of a few FM clusters embedded in the paramagnetic background in the temperature range from 210 K to 270 K. It is believed that the magnetic field can enlarge the FM clusters a little, but they are not large enough to connect each other to create metallic regime. Furthermore, it is regarded that the strain from lattice distortions has a large effect on CO formation, and CO can induces a lattice distortion through the electron-lattice coupling manifested by the Jahn–Teller effect.[15,16] Fernandez-Diaz *et al.*[17] have studied the resistivity, magnetic susceptibility, and specific heat of the compound LCMO from 1.5 K to 400 K, and found a charge-ordering transition taking place at around 270 K. At 250 K, the change of CO state is obvious, and an antiferromagnetic order is mainly established at 170 K. A promising description of the mechanism of those transport properties is the phase separation scenario, suggesting that the manganite has a mixed-phase state property with intrinsic inhomogeneity.

In this Letter, we present a calculation by using BFT algorithm[12,13,18] to simulate the resistivity of LCMO according to the random resistor network model, based on phase separation scenario. The theoretical simulation results are in quantitative agreement with the experimental data reported by Fernandez-Diaz *et al.*[17] in the temperature range from 80 K to 350 K, showing a transition from a high-temperature PM/CDO insulating state embedded with a few FM metallic clusters to a low-temperature AF/CO insulating state. It is believed that CO state formation with the decrease of the temperature suppresses FM clusters, so that FM clusters disappear

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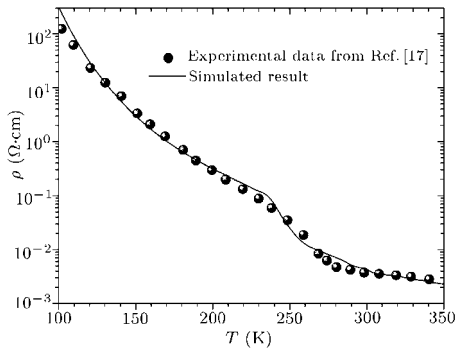

Fig. 1. Simulated T-dependent resistivity (solid line) of LCMO compared with the experimental data (circles) obtained from Ref. [17] with denary coordinate in the y axis.

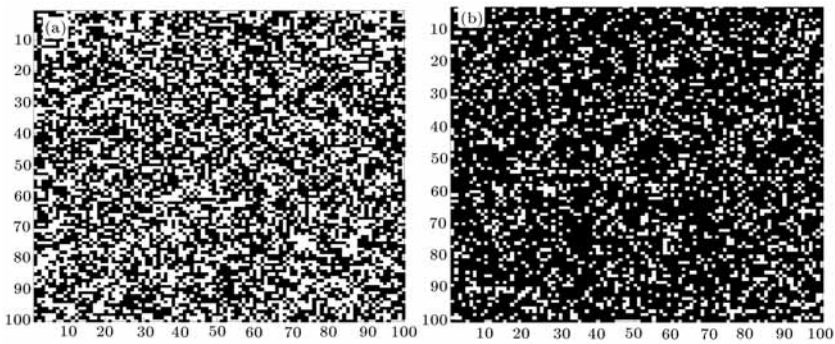

Fig. 2. Simulated result of the AF/CO (black) and PM/CDO (white) of LCMO for (a) at $\sim 250\,\mathrm{K}$ and for (b) at $\sim 170\,\mathrm{K}$, in the mixed-phase description on 100×100 clusters.

soon. All these results indicate that the coexistence of phases is crucial for the behaviour of the transport in LCMO.

The main concept of the random network model in the system, a two-dimensional $N \times N$ matrix, is composed of two phases with different conductive properties. One is the PM/CDO insulating region, the other is the AF/CO insulating region. Because the number of the FM clusters which are almost engulfed by the surrounding PM/CDO insulating state is very small, we can presume that the FM clusters embedded in the paramagnetic insulating state background as impurity enhances the electrons scattering slightly in PM/CDO insulating state. In order to simplify the model, it is reasonable to assume that the FM clusters are not of independent phase, and they only enhance the resistivity of PM/CDO insulating state. A quantity p, defined as the ratio of the number of AF/CO lattices to the number of the total lattices, represents the fraction of AF/CO sites ($0 \leq p \leq 1$), and decreases with the increasing temperature T. At low temperature, p is equal to 1 due to the complete antiferromagnetic charge-ordered state, and equal to 0 at high temperature due to the complete paramagnetic charge-disordered state. For the intermediate temperature range, p is between 1 and 0, representing the coexistence of PM/CDO and AF/CO states. Hence, p is a T-dependent function, and this T-dependent variation of AF/CO component is not linear. With the decreasing T, AF/CO phase starts to appear, and its fraction increases slowly. However, the sample is still mainly occupied by the PM/CDO state. Only below the temperature at which AF/CO clusters begin to percolate, the fraction of AF/CO phase increases rapidly, and the AF/CO state is dominant in the sample. The variation of AF/CO fraction on the temperature dependence described above is just what happened in the phase transition process observed in Ref. [15]. Therefore, for simplification, it is reasonable to assume that the fraction of AF/CO sites, p, is a Fermi distribution function of temperature T as follows:

$$p = \{1 + B \exp[A(T - T_{\mathrm{AF/CO}})]\}^{-1},$$

where $T_{\mathrm{AF/CO}}$ denote the temperature of AF/CO states at which current starts to percolate. A and B are the constants depending only on the material.

We assume that the PM/CDO insulating regions have the resistance R_{PI} (T), and the AF/CO insulating regions have the resistance R_{AI} (T). The total effective resistance R_{eff} is determined by the parallel connection of R_{PI} (T) and R_{AI} (T). To calculate R_{PI} (T) and R_{AI} (T), we assume $\rho_{pi}(T) = \rho_{pi0} \exp[E_{p0}/(k_B T)]$ and $\rho_{ai}(T) = \rho_{ai0} \exp[E_{a0}/(k_B T)]$ are the T-dependent resistivities for each PM/CDO site and AF/CO site, respectively.[12−14] Here ρ_{pi0} and E_{p0} are the high-T residual resistivity and activation energy for the PM/CDO state respectively; Correspondingly, ρ_{ai0} and E_{a0} are those for the AF/CO state, respectively; k_B represents the Boltzmann constant. These parameters (ρ_{pi0}, E_{p0}/k_B) and ($\rho_{ai0}, E_{a0}/k_B$) can be fitted by the experimental data in the high-T and low-T experimental data, respectively. Considering the effect of the FM clusters, the activation energy E_{p0} is enhanced due to the reason mentioned above. Using the BFT algorithm,[12,13,18] the path lengths of the PM/CDO and AF/CO domains are found, then the resistances $R_{\mathrm{PI}}(T)$ and $R_{\mathrm{AI}}(T)$ are derived from the obtained path lengths, respectively. Finally, the effective ρ can be obtained according to the size of the sample.

The experimental T-dependent resistivity curves of LCMO obtained from Ref. [17] with magnetic field $H = 9\,\mathrm{T}$ are presented in Fig. 1 with the logarithm coordinate in the y-axis, where the circle symbols denote the experimental data observed in warming process. Under an external magnetic field of 9 T, the CO state is stable.[17] Based on the model and method mentioned above, we have used the corresponding parameters listed in Table 1. The simulated results of

Table 1. Parameters used in simulation for LCMO.

A	B	ρ_{pi0} (Ωcm)	ρ_{ai0} (Ωcm)	E_{p0}/k_B (K)	E_{a0}/k_B (K)	$T_{\mathrm{AF/CO}}$ (K)
0.0128	0.773	0.0001	0.00019	1009.13948	1421.68811	250

the T-dependent resistivity are given in Fig. 1 by the solid line.

Based on our above-mentioned model, we can find that the simulated curve in Fig. 1 is in good agreement with the experimental data, exhibiting a transition of a PM/CDO insulating phase embedded with a few FM metallic clusters to an AF/CO insulating phase. It is found that the AF/CO clusters are initially formed with decreasing temperature in paramagnetic charge disordering phase, and the AF/CO fraction increases as the temperature decreases. Then the AF/CO clusters percolate at $T_{\mathrm{AF/CO}} \sim 250$ K, and the AF/CO state is mainly established at low temperature. It should be pointed out that the effect of FM metallic clusters is weak due to the very small number of clusters. Moreover, the AF/CO state formation, the lattice distortion and the super-exchange driven antiferromagnetic spin fluctuations with decreasing temperature all can restrict the hopping of e_g electrons, which induces ferromagnetic correlations through the double-exchange mechanism, so that the FM clusters disappear soon.[15–17] Under an external magnetic field of 9 T for LCMO, the CO state is stable. These are very different from the MIT behaviour of many compounds such as $La_{1-x}Ca_xMnO_3$ at $\sim 0.2 < x < 0.5$.[1,9,10,19,20] Considering the changes in the corresponding parameters values of the resistivities for each PM/CDO site and AF/CO site, it is found that ρ_{pi0} and E_{p0} are smaller than ρ_{ai0} and E_{a0}, respectively. Thus, we can conclude that the electrical transport property in the PM/CDO state is entirely different from that in the AF/CO state. The free electrons in the AF/CO states are more difficult to move than those in the PM/CDO states, although the resistivity of the two states are of similar formula. All these results indicate that intrinsic inhomogeneity plays a crucial role in the electrical transport property in the formation of the coexisting of CDO and CO insulating phases.

To further understand the coexistence of the phases and the percolative conductive behaviour from the phenomenological view, a simulated result is demonstrated in Fig. 2 at $\sim$ 250 K and at $\sim$ 170 K on a 100×100 matrix, respectively. The number of AF/CO squares is determined by the T-dependent p. The position of the AF/CO square on the 100×100 network is random. With the decrease of T, PM/CDO squares are randomly selected to be converted into AF/CO squares. The small dark squares represent the AF/CO sites and the white ones denote the PM/CDO areas. We can find that the number of AF/CO sites is only slightly more than that of PM/CDO sites at 250 K. However, only a few PM/CDO domains near a AF/CO state can be observed at 170 K.

In summary, by using the random network model based on the phase separation scenario, we have simulated the resistivity of the LCMO. It is found that the number of AF/CO clusters increases with the decreasing temperature, and the clusters start to connect to each other around 250 K. The agreement between the simulated results and experimental data indicates that our model is appropriate for describing transport behaviour of the material with phase separation and phase separation indeed exists in LCMO, and that the intrinsic inhomogeneity plays a crucial role in the electrical conductivity.

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References

[1] Jin S, Tiefel T H, Mocormack M, Fastnacht R A, Ramesh R and Chen L H 1994 *Science* **264** 413
[2] Lu H B et al 2004 *Chin. Phys. Lett.* **21** 2308
Lu H B et al 2004*Appl. Phys. Lett.* **84** 5007
Duan P et al 2004 *Appl. Phys. Lett.* **84** 4741
[3] Guo H Z et al 2004 *Appl. Phys. Lett.* **85** 3172
Zhao K et al 2005 *Chin. Phys.* **14** 420
Zhao K et al 2005 *J. Rare Earth* 23 526
[4] Jin K J et al 2005 *Phys. Rev.* B **71** 184428
Lu H B et al 2005 *Appl. Phys. Lett.* **86** 241915
Zhou Q L et al 2005 *Phys. Rev.* B **72** 224439
[5] Tian H F et al 2005 *Appl. Phys. Lett.* **87** 164102
Zhou Q L et al 2005 *Appl. Phys. Lett.* **87** 172510
Lu H B et al 2005 *Appl. Phys. Lett.* **86** 32502
[6] Jin K J et al 2005 *Sci. Technol. Adv. Mater.* **6** 833
He M et al 2005 *Sci. Chin. G* **35** 625
Huang Y H et al 2005 *Sci. Chin. G* **48** 381
Zhao K et al 2005 *Appl. Phys. Lett.* **86** 221917
[7] Zhao K et al 2006 *Appl. Phys. Lett.* **88** 141914
Jin K J et al 2006 *J. Magn. Magn. Mater.* (in press)
Huang Y H et al 2006 *Chin. Phys. Lett.* **23** 982
[8] Zhao K et al 2006 *Physica* B **373**72
Zhou Q L et al 2006 *Phys. Lett.* A **350** 283
Guan D Y et al 2006 *Chin. Phys. Lett.* **23** 460
[9] Millis A J 1998 *Nature* **392** 147
[10] Dagotto E, Takashi Hotta and Adriana Moreo 2001 *Phys. Rep.* **344** 1
[11] Mayr M et al 2001 *Phys. Rev. Lett.* **86** 135
[12] Zhou Q L et al 2005 *Chin. Phys. Lett.* **22** 1749
[13] Guan D Y et al 2005 *Phys. Status Solidi* A **202** 2776
[14] Zhou H D et al 2002 *Solid. State Commun.* **122** 507
[15] Tao J and Zuo J M 2004 *Phys. Rev.* B **69** 180404
[16] Loudon J C and Midgley P A 2005 *Phys. Rev.* B **71** 220408
[17] Fernandez-Diaz M T, Martinez J L, Alonso J M and Herrero E 1999 *Phys. Rev.* B **59** 1277
[18] Akiena A 1990 *Implementing Discrete Mathematics: Combinatorics and Graph Theory with Mathematica* (San Francisco: Addison-Wesley)
[19] Pradhan A k et al 2001 *Appl. Phys. Lett.* **79** 506
[20] Rao C N and Raveau B 1998 *Colossal Magnetoresistance, Charge Ordering and Related Properties of Manganese Oxides* (Singapore: World Scientific)