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Resistivity and critical temperature of $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$*

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An empirical formula of the critical temperature that is concentration dependent for polycrystalline $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$ is presented in this paper. With this formula, the temperature dependence of resistance is simulated for various values of x by using the random resistor network model and the Monte Carlo method. The hysteresis effect in $\rho - T$ curves is reasonably explained. The simulation results are in good agreement with the relevant experimental measurements.

Keywords: Monte Carlo simulation method, colossal magnetoresistance, phase separation, hysteresis effect

PACC: 7540, 7530V, 7530, 7130

1. Introduction

The ABO_3-type doped manganite perovskites, namely $R_{1-x}D_xMnO_3$ (where R denotes rare earth metal and D represents alkaline earth metal), have been extensively investigated in recent years because of their colossal magnetoresistance (CMR) effect.[1,2] The simultaneous appearance of ferromagnetic (FM) and metallic behaviours in these systems is attributed to the double exchange (DE) interaction between pairs of Mn^{3+} and Mn^{4+} ions.[3] Substitution of cations with different sizes for the rare earth sites results in a lattice distortion that influences the FM double exchange and the antiferromagnetic (AFM) superexchange interactions. Recently it has been demonstrated that microscopic phase separation plays an essential role in the physics of the rare earth manganites.[4−6] Essentially, the phase separation can result in the apparent percolative feature of the insulator-metal transition when the transition comes from the charge-ordered insulating to the FM metallic state. Here one observes the largest change in resistivity (more than 4 orders of magnitude) and, therefore, the largest magnetoresistance. Even a small distortion of the atomic arrangement, hence the domain structure, can induce the percolation.

In spite of the large amount of work devoted to the manganites, the microscopic nature of the phase-separated state has not been well understood. For example, the electronic properties of the constituent phases as well as their volume fraction and spatial distributions in the sample remain unclear. To address these basic problems, the microscopic structure of manganites must be characterized in detail. In this regard field-dependent resistivity and the Seebeck coefficient play an important role.

The $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$-type system, where La is partially substituted for Pr that has a smaller ionic radius, turns out to be one of the most convenient systems for studying the phase separation behaviour. The Pr-rich composition corresponds to a very stable charge ordered AFM phase, whereas the compositions with high La content are FM metals. Recently, Patil and Bhagat investigated this type of compound and measured the variation in dc resistivity with temperature, showing that there exists a thermal hysteresis effect when $x > 0.4$.[7]

In this paper, we give an empirical formula of the critical temperature which is concentration dependent for this type of compound. With this formula we simulate the temperature dependence of resistance in $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$ for various values of x by using the random resistor network model and the Monte Carlo method that has been extensively used in colossal magnetoresistance and strongly correlated

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systems.[8,9] We also give a reasonable explanation to the hysteresis effect in ρ–T curves. The simulation results are in good agreement with the relevant experimental measurements.

2. Model and simulation

It has been commonly believed that the perovskite-related manganites can accomplish a dynamic phase segregation.[10] Because of the phase segregation, it is likely for the optimal manganese perovskites that FM domains exist within the PM matrix in the temperature range $T > T_c$, while PM regions exist in the FM background in the temperature range $T < T_c$. Experimental evidence for the existence of short-range FM domains in the PM background can be found in experiments such as electron paramagnetic resonance[11] and small-angle neutron scattering measurements.[12] Experiments such as scanning tunnelling spectroscopy,[13] magnetic force microscope[14] and electron microscopy[15] measurements, on the other hand, directly confirm the coexistence of FM domains and PM regions below T_c. According to the DE mechanism,[3] the charge carriers with the same spin orientations can move easily; therefore, the electrical conductivity is metallic in the FM state, whereas the electrical conductivity is insulating in the PM state because of the disordered scattering from charge carriers with different spin orientations. It is therefore reasonable to assume that the sample is a phase-separated-into-mixture of FM metallic domains and PM insulating regions.

Although there have been some attempts in the literature to calculate dc resistivity ρ_{dc}, typically a variety of approximations that are not under control has been employed. In fact, some of the coexistent clusters found in experiments, which were in the micrometre size range, strongly suggest that a fully microscopic approach to the problem should very likely fail in a computational analysis. It is clear that a more phenomenological approach is needed.

On the basis of the above consideration, Mayr *et al* [16,17] adopted a random resistor network model and other approximations. However, all these simulations involve little in hysteresis effect in $\rho - T$ curves. As a phenomenological model, the total resistance is represented as the sum of the contribution from the PM insulating phase and that from the FM metallic phase. It is shown that this phenomenological model can quantitatively explain experimental observations on the optimal manganese perovskites. However, FM metallic and PM insulating regions are assumed to be electrically connected in series. Clearly, the connection in an actual system should be much more complex than this assumed form. The actual situation should be that FM metallic and PM insulating regions randomly fill the space of sample. In order to create such a distribution, the Monte Carlo method is used.

We assume that the sample is composed of dense grains of PM insulating particles and of FM metallic particles packed together so that the particle centres occupy the nodes of a network lattice. Between each pair of particles in the network lattice a current flows through a 'bond resistance'. The bond resistivity is determined from the kinds of particles residing at sites i and j. Between each pair of particles of the same kind (FM metal or PM insulator), the bond resistivity between sites i and j is determined by $\rho_{ij} = \rho_I$ (or $\rho_{ij} = \rho_M$). If the two sites are occupied by different kinds of particles, i.e. an FM metallic particle at site i and a PM insulting particle at site j, the bond resistivity between sites i and j is determined by $\rho_{ij} = \dfrac{\rho_I \rho_M}{\rho_I + \rho_M}$.The kinds of particles residing at sites i and j depend on the value of a computer-generated random number. If this number is smaller than the chosen number density (P) of FM metallic particles, the site is assigned to an FM metallic particle, and otherwise the site is taken to be occupied by a PM insulating particle. Through this process, the random resistor network can then be constructed. Application of the Kirchhoff current law to each mesh, we constitute a set of equations that may be solved numerically.

To perform the Monte Carlo simulation, we first need to know three quantities: ρ_I, ρ_M and P, which appear in the random-resistor-network model. Both ρ_M and ρ_I can be approximated as $\rho_M = bT^2$ and $\rho_I = a \times \exp(\dfrac{c}{t})$ respectively. Since P represents the number density of FM metallic particles, it should be proportional to the reduced magnetization M. we therefore approximate P as M that can be determined by the mean-field theory. According to this theory, M can be expressed in terms of solutions of the mean-field self-consistent equation[18]

$$M(h,t) = B_J(\frac{3J}{J+1}\frac{M+h}{t}),$$

where $B_J(x)$ is the Brillouin function determined by

$$B_J(x) = \frac{2J+1}{2J}\mathrm{cotanh}\left(\frac{(2J+1)x}{2J}\right) - \frac{1}{2J}\mathrm{cotanh}\left(\frac{x}{2J}\right).$$

Here J is the angular momentum quantum number, $t(\sim T/T_c)$ is the normalized temperature, $h(\sim H/H_e)$ is the normalized magnetic field and H_e is the saturation exchange field. For the compound $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$, we assume that the field is zero, i.e. $h = 0$, and the constant J is equal to 1.85. Constants a, b and c are given in the following table:

x	0.4	0.5	0.6	0.7	0.8
a	1.98×10^{-2}	9.16×10^{-3}	7.66×10^{-3}	4.59×10^{-3}	1.24×10^{-3}
b	3.83×10^{-5}	4.16×10^{-5}	5.88×10^{-4}	2.08×10^{-3}	3.57×10^{-3}
c	1049	1241	1286	1452	1500

For the parameter T_c , we assume an empirical formula in the following form:

$$T_c^{\pm} = \sum_{i=0}^{6} a_i x^i \pm \theta(x - x_c) \sum_{j=0}^{3} b_j x^j,$$

where $T_c^{\pm}$ are the critical temperatures in heating(+) and cooling(-) processes respectively, x_c is the critical value of x at which the hysteresis phenomenon starts to appear. For $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$, $4 < x_c < 5$, $a_0 = 250.000$, $a_1 = -1209.196$, $a_2 = 13867.574$, $a_3 = -61475.446$, $a_4 = 127907.366$, $a_5 = -127678.571$, $a_6 = 49144.345$, $b_0 = -41.5$, $b_1 = 193.333$, $b_2 = -300$ and $b_3 = 166.667$, the simulated ρ – T curves are shown in Fig.1.

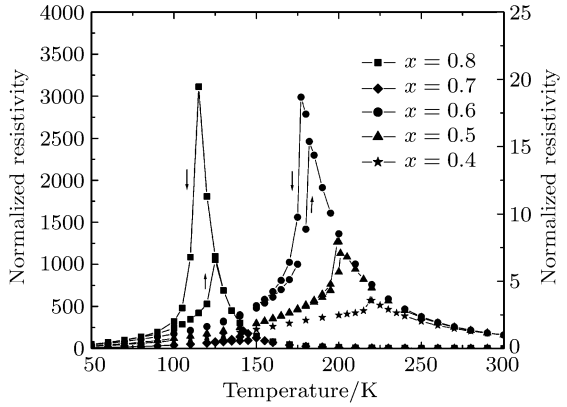


Fig.1. The curves of resistivity vs temperature for various concentrations x, showing that thermal hysteresis appears when $x > 0.4$. The upward arrows show the curves corresponding to the heating process.

3. Discussion

Figure 1 shows the temperature dependence of the resistivity in both heating and cooling processes. At temperatures below T_c, the resistivity increases with the increase of temperature, corresponding to the metallic behaviour. By further increasing the temperature it rapidly reaches a maximum valve at T_c, and then decreases in the region $T > T_c$, indicating the first-order transition from metal to insulator. A general phenomenon associated with the first-order transition is the presence of hysteresis in cycling through the transition and the relevant effects of superheating and supercooling. Thermodynamically the transition should occur at T_c. In the cooling process, the high temperature phase is stable against small fluctuation until a temperature T^*, the limiting temperature of metastability of supercooling. Between T_c and T^*, the low-temperature ordered phase will occur only if there exists a sufficiently large fluctuation. Below T^* the system is unstable against infinitesimal fluctuations to the low-temperature ordered phase. Thus, on cooling, the actual transition to the low-temperature phase will occur at some temperature T^- between T_c and T^*, depending on the particular sample and the experimental condition. Similar arguments can apply to the heating process and the actual transition temperature T^+. This is a main reason why we suggest that the empirical formula of the critical temperature is in such a form as mentioned above. It is clear that our simulation results are in good agreement with the experimental measurements. In the simulation as mentioned above, we have used a (uncorrelated) random resistor network model. We also investigate the possibility that the correlated occupation[19] of metallic clusters could be used to explain the hysteresis of $(La_{1-x}Pr_x)_{0.7}Ca_{0.3}MnO_3$ based on a correlated ran-

dom resistor network model in which the possibility of adding new metallic (or insulating) clusters to a certain site is larger when there exist already metal-occupied sites adjacent to it. We do not find that this method based on the correlated occupation is suited to explain this kind of hysteresis, although it is plausible to be used to explain the 'overshot' hysteresis in Ref.[19].

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