Simulation of Anisotropic Resistivity for Mixed-Phase Manganite $La_{2/3}Ca_{1/3}MnO_3$ Thin Films *

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We utilize the random network model based on phase separation scenario to simulate the conductive behaviour and anisotropic characteristics of resistivity for $La_{2/3}Ca_{1/3}MnO_3$ (LCMO) thin films. The simulated results agree well with our experimental data, showing a metal-to-insulator transition from a high-T paramagnetic (PM) insulating phase to a low-T ferromagnetic (FM) metallic phase in both the untilted film deposited on a (001) $SrTiO_3$ (STO) substrate and the tilted film grown on a vicinal cut STO substrate. It is found that the resistivity of the tilted sample is higher than that of the untilted one, displaying prominent anisotropic characteristics. The studies reveal that the tilting not only decreases the conduction of the FM domains, but also increases the activation energy of the PM regions, inducing the enhancement of resistivity. All those results suggest that the intrinsic inhomogeneity in the phase separation system plays a significant role in the electrical conductivity and the resistive anisotropy is related to the structure of the crystal lattice.

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Mixed-valence manganite oxides, in particular the lanthanum manganites, have attracted wide attention since the observation of colossal magnetoresistance (CMR) effect and the related intriguing phenomena.[1] Without hole doping, parent compound $LaMnO_3$ (LMO) is insulating at all temperatures. However, when the La^{3+} ion in LMO is substituted by a divalent cation as in $La_{1-x}A_xMnO_3$ (A = Ca, Sr or Ba), the material is generally a ferromagnetic (FM) metal below T_c and a paramagnetic (PM) insulator above T_c at $\sim 0.2 < x < 0.5$.[2] Furthermore, due to the distortion of the lattice, the doped LMO compound is not the ideal cubic perovskite structure anymore, presenting anisotropic characteristics. Anisotropic magnetoresistance (AMR), as one of the important properties, has been widely investigated for novel applications in new devices.[3] The anisotropic resistivity has been reported on double-layered manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$ and LCMO, showing the obvious anisotropy of resistivities measured along the c- and a-axes.[4−6] However, few studies have been carried out on the anisotropic resistivity in the tilted thin films. In addition, the transport properties remain unsolved since theoretical calculation of resistivity is notoriously difficult. Hence, it is necessary to understand the true physics of the metal-to-insulator transition (MIT) and the anisotropic resistivity. The double-exchange model[7] and the Jahn–Teller phonon effect[8] are not sufficient in explaining the phase transition and the enormous magnitude of CMR.[9] A prospective description is the percolative conduction in the phase separation system, which means that one kind of phases exists in the background of other phase or phases. Fäth *et al.*[10] has observed the coexistence of PM and FM regions below T_c by the scanning tunnelling spectroscopy. Based on the phase separation scenario, Mayr *et al.*[11] used a random resistor network to simulate the T-dependent resistivity and obtained a qualitative result in agreement with the experimental results.

In this Letter, we present a calculation method for the random network model to simulate the anisotropic resistivity based on the phase separation scenario combined with the crystal structure for $La_{2/3}Ca_{1/3}MnO_3$. The theoretical results give excellent quantitative agreement with our experimental data in the temperature range from 5 K to 310 K, showing an MIT in the untilted and tilted thin films. By considering the influence of crystal structure on anisotropic resistivity, we use two different ways to simulate the resistivity of the tilted film and obtain the same result. In addition, it is revealed that the tilting can increase the resistivity of both the FM and PM states to enhance the effective resistivity of the sample. The studies indicate that the coexistence of the phases is crucial for understanding the physics of manganites and the crystal structure plays an important role in anisotropic resistivity.

Figure 1 shows a schematic diagram of the percolative framework. Two-dimensional $N \times N$ square matrix shown in Fig. 1(a) is used to simulate the trans-

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port behaviour. The lattice spacing of this network is much larger than the microscopical Mn–Mn distance, comparable to the macroscopical FM (gray) or PM (white) domain size. In such a matrix, the conductivity is metallic within the FM regions due to the double exchange, but insulating in the PM regions because of the disorder scattering. Thus, it is assumed that there are two types of components with different conductive properties in the system. A quantity f, defined as the ratio of the number of FM lattices to the number of total lattices, is used to represent the fraction of FM metallic sites. At each site in this network, either a metallic or insulating resistance is located randomly and the total fraction of metallic component is f $(0 \leq f \leq 1)$. Obviously, f is equal to 1 at extreme low temperature due to the complete ferromagnetism and equal to 0 at high temperature due to the complete paramagnetism. For the intermediate temperature range, f is between 1 and 0, representing the coexistence of FM and PM states. Hence, a T-dependent metallic fraction $f(T)$ is needed. It should be noticed that f must decrease with the increasing T and should change rapidly near T_c,[11] similar to the T-dependent magnetization. For simplification, a reverse sigmoid function, such as Fermi distribution function, is used to describe the change of f induced by the temperature.

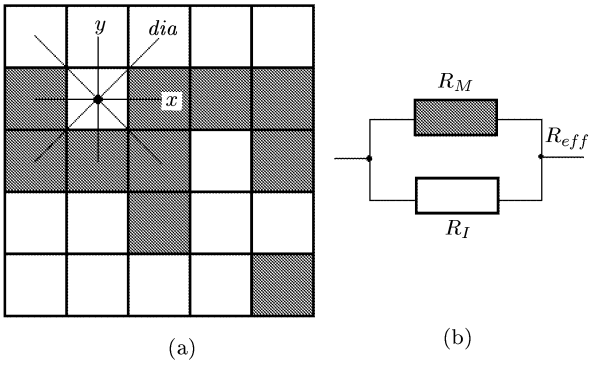

Fig. 1. (a) Schematic representation of the mixed-phase state about the distribution of the FM metallic (gray) and PM insulating (white) domains. (b) Two-resistance model for calculating the effective resistance $R_{\rm eff}$ from the parallel connection of metallic R_M and insulating R_I resistances.

We divide the system into two parts, as illustrated in Fig. 1(b). One is the FM metallic region with the resistance $R_M(T)$ and the other is the PM insulating region with the resistance $R_I(T)$. The total effective resistance $R_{\rm eff}$ is determined by the parallel connection of $R_M(T)$ and $R_I(T)$. To calculate the resistance R_M and R_I, we assume $\rho_m(T) = \rho_{m0}+\rho_{m1}T^2+\rho_{m2}T^{4.5}$ and $\rho_i(T) = \rho_{i0}\exp[E_0/(k_BT)]$ are the T-dependent resistivities for each FM site and each PM site, respectively.[12,13] In the FM metallic state, ρ_{m0} is the residual resistivity at $T \sim 0\,$K, the T^2 term indicates the electron scattering[14] with the coefficient ρ_{m1}, and the $T^{4.5}$ term denotes the magnon scattering involving the phonon scattering[15] with the coefficient ρ_{m2}. In the PM insulating state, ρ_{i0} is the high-T residual resistivity, E_0 is the activation energy, and k_B represents the Boltzmann constant. These parameters $(\rho_{m0}, \rho_{m1}, \rho_{m2})$ and $(\rho_{i0}, E_0/k_B)$ can be fitted from the experimental data in the low-T and high-T experimental data, respectively.

By using the Breadth-First Traversal algorithm,[16] which explores all nodes adjacent to the current node before moving on and can be used to compute the shortest path from the source to all reachable nodes and the shortest-path distances, the path lengths of the metallic and insulating domains are found. Then the resistances R_M and R_I are derived from the obtained path lengths, respectively. Finally, the effective resistivity ρ can be obtained according to the size of the sample.

To explain the conductive behaviour of LCMO, here we present our experimental data of the untilted and tilted samples in the temperature range from 5 K to 310 K, which will be published elsewhere. The results show the electrical resistivities undergo MITs and are indeed anisotropic with maximum resistivity ratio (ρ_tilted)/(ρ_untilted) of $\sim$ 2.4 around 235 K, even if the tilted angle is only 10°. The untilted and tilted LCMO thin films were deposited by laser molecular-beam epitaxy (laser MBE)[17] on different STO substrates under the same deposition condition. The growth on the untilted (001) substrate will make film with the c-axis perpendicular to the plane. The STO substrate that is intentionally miscut by 10° towards the [010] direction will keep the c-axis of LCMO oblique to the plane. Here, the LCMO orthorhombic structure is referred with the *Pbmm* rotation, with $a \approx b$ the short axis and c the larger axis.[6] The films were structurally characterized by x-ray diffraction, showing excellent crystalline qualities. The electrical resistivity measurement was performed with the standard four-point technique.

Based on the model and method mentioned above, the simulated result of the T-dependent resistivity is obtained on 100×100 clusters for the untilted sample, as presented in Fig. 2. The corresponding experimental data (circles) are also given in this figure, showing the T-dependent resistivity undergoes an MIT with the increasing temperature. The MIT temperature T_{MI} is about 260 K, nearly equal to $T_c \sim 267\,$K of the sample. The simulated curve (solid line) shows a good agreement with the experimental result, exhibiting the changes from low-T behaviour of $d\rho/dT > 0$ to high-T behaviour of $d\rho/dT < 0$. Below T_c the long-range magnetic ordering allows the transfer of the charge, the behaviour being metallic-like. The semiconducting-like behaviour above T_c is associated

with the existence of a polaronic regime.[18] It can be seen that the model not only results in the MIT, but also yields quantitative fits to the experimental data over the whole studied temperature range.

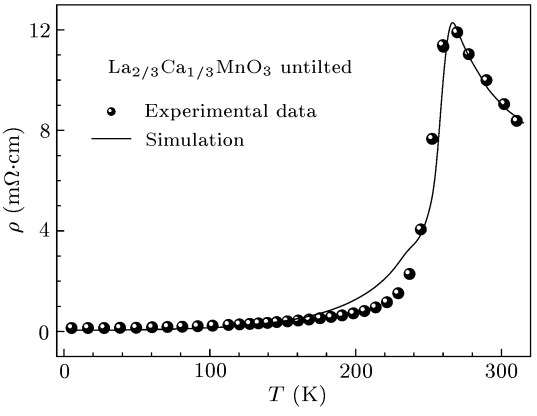

Fig. 2. Calculated T-dependent resistivity (solid line) of LCMO without tilting on a 100 × 100 matrix compared with the experimental data (circles).

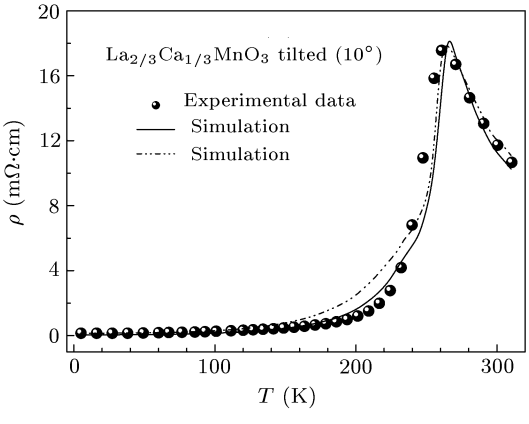

Fig. 3. Simulated results of resistivity for 10°-tilted LCMO with the corresponding experimental data (circles). The solid line represents the simulated result obtained by only adjusting weights of directions. The dashed line represents the simulated result obtained by only adjusting resistivity parameters.

Now we consider the tilting influence on the resistivity of LCMO. We have simulated the transport behaviour using two different methods. On the other hand, we consider the resistivity of each square, where the current flows, is also proportional to the weight of path directions. In fact, the metallic and insulating domains are not exact squares and percolation will occur when two squares have a shared edge or point, that is, we search paths in eight directions for each square, as shown in Fig. 1(a). If the sample is untilted with ab-axis in the film plane, those eight directions are equivalent, displaying isotropy. However, when the sample is tilted, the path directions will exhibit anisotropy, similar to the condition that there are some c-axis components in the film plane along the tilting direction. For simplification, we regard the weight of x direction is unchanged and equal to 1. However, the weights of y and diagonal directions are different from that of x direction and satisfy the relation $x^2 + y^2 = d^2$. Keeping other parameters unchanged, we only adjust the weights of the y and diagonal directions and then obtain the simulated result, as shown by the solid line in Fig. 3. Thereinto, the weight for the y direction is 2 and that for the diagonal direction is $\sqrt{5}$. It can be seen easily that MIT still exists and the MIT temperature seems unchanged. The excellent agreement between the simulated results and experimental data indicates that tilting can enhance the resistivities of those squares and can induce a higher effective resistivity compared with the untilted one.

On the other hand, instead of considering the weight of path directions, we only adjust the coefficients of the resistivities for FM and PM sites. Distinctly, there is also a good agreement between the experimental data and the simulated result (the dashed line in Fig. 3), showing a higher resistivity after tilting. The corresponding parameters used are listed in Table 1 for the untilted and tilted samples. There are some changes in the values of these parameters. Firstly, ρ_{m0}, ρ_{m1} and ρ_{m2} are higher than those without tilting. This can be understood as due to the reason that the tilting increases the spin scattering in conduction carriers and decreases the electron hopping probability due to the change of Mn-O bond in the plane.[6] Secondly, the value of E_0/k_B is increased, indicating that the tilting can enhance the activation energy. For manganites with relatively smaller cation-like Ca, the paramagnetic phase is generally insulating with a activation energy E_0 and there is an interesting dependence of E_0 on the Mn–O distance wherein it increases with the Mn–O distance.[2] Thus, the increase of Mn-O bond length along the tilting direction would lead to an increase of the resistivity. Finally, ρ_{i0} remains unchanged and this is also reasonable because the tilting may not reduce the resistivity of the PM components at very high temperature. From the simulated results, we find that these two methods, changing the weight of path direction and adjusting the coefficients of resistivity formula, can yield to the same result for the resistivity of the tilted sample. Those results further verify that our consideration is reasonable about the tilting influence on the resistivity.

Table 1. Parameters used in the simulation for untilted (0°) and tilted (10°) LCMO thin films, respectively.

θ (deg)	ρ_{m0} (mΩ·cm)	ρ_{m1} (mΩ·cm·K^{-2})	ρ_{m2} (mΩ·cm·K^{-4})	ρ_{i0} (mΩ·cm)	E_0/k_B (K)
0	0.135	7.0×10^{-6}	2.8×10^{-11}	5	100
10	0.146	8.0×10^{-6}	6.4×10^{-11}	5	200

In summary, we have simulated the T-dependent resistivity for untilted and tilted LCMO thin films

by using the random network model based on the phase separation scenario. The results indicate that the present model can quantitatively explain conductive transport behaviour observed in our experiments, showing an MIT of resistivity and exhibiting remarkable anisotropic resistivity for LCMO. We find that the tilting has a significant influence on the conductive behaviour of the manganites and can induce electrical resistivity anisotropy due to the change of Mn–O bond length. The tilting can increase the resistivity of the FM metallic domains by enhancing the scattering, as well as the PM insulating regions by enhancing the activation energy, to raise the effective resistivity of the sample. The agreement between the simulated results and our experimental data indicates our model is appropriate for describing the electrical conductivity of the manganites with the intrinsic inhomogeneities in the form of coexisting competing phases. It can be concluded that the crystal structure plays an important role in anisotropy of resistivity.

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