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Thermal Hysteresis in $La_{2/3}$ $Ca_{1/3}$ MnO_3 Films Grown on Tilted $SrTiO_3$ Substrates and Influence of External Magnetic Field *

GUAN Dong-Yi(关东仪)[1], ZHOU Qing-Li(周庆莉)[1], ZHAO Kun(赵昆)[1], SUN Jia-Xing(孙嘉兴)[2],
JIN Kui-Juan(金奎娟)[1**], CHEN Zheng-Hao(陈正豪)[1], LU Hui-Bin(吕惠宾)[1], YANG Guo-Zhen(杨国桢)[1]
[1]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Science, PO Box 603, Beijing 100080*
[2]*Department of Physics, Liaoning University, Shenyang 110036*

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Thermal hysteresis in resisitivity of $La_{2/3}Ca_{1/3}MnO_3$ thin films grown on tilted $SrTiO_3$ substrates is simulated by using the random network model on the basis of mixed-phase percolation between metallic and insulating domains. The metallic-insulating transition temperatures during the warming process are lower than those during the cooling process due to the difference in fraction of metallic domains between warming and cooling process. With an external magnetic field, the metallic-insulating transition temperatures shift to a higher value and the resistivities are reduced. The excellent agreement between the simulation and the experimental data further verifies that phase separation plays a crucial role in the transport process of $La_{2/3}Ca_{1/3}MnO_3$ thin films.

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The manganites usually have a metal-to-insulator transition (MIT) accompanied by a simultaneous phase-to-phase transition at almost the same temperature T_c.[1-3] During warming and cooling processes, the resistivity behaves thermal hysteresis. In this Letter, we simulate the resistivity of $La_{2/3}Ca_{1/3}MnO_3$ films grown on tilted $SrTiO_3$ substrates in a thermal cycling by using the random network model based on the prevailing phase separation scenario and study the influence of external magnetic field on their transport behaviour. The good results obtained from the simulations resemble the ones observed in the experiments.

The main concept of the random network model is that the system, a two-dimensional $N \times N$ matrix, is composed of two kinds of sites with different conductive properties that are metallic and insulating. A quantity f, defined as the ratio of the number of FM *lattices* to the number of total lattices, represents the fraction of FM metallic sites ($0 \leq f \leq 1$). Obviously, the fraction of PM insulating sites is $(1 - f)$. We assume $\rho_m(T) = \rho_{m0} + \rho_{m1}T^2 + \rho_{m2}T^{4.5}$ and $\rho_i(T) =_{i0} \exp[E_0/(k_BT)]$ are the T-dependent resistivities for each FM site and PM site, respectively. Here ρ_{m0} is the residual resistivity at $T \sim 0\,K$, the T^2 term indicates the electron scattering[4] with the coefficient ρ_{m1}, and the $T^{4.5}$ term denotes the magnon scattering involving the phonon scattering[5] with the coefficient ρ_{m2}. The coefficient ρ_{i0} is the high-T residual resistivity, E_0 is the activation energy, and k_B represents the Boltzmann constant.[6] The resistance of the metallic $R_M(T)$ or insulating region $R_M(T)$ is respectively derived as the product of the resistivity of $\rho_m(T)$ or $\rho_i(T)$ and the shortest length of the connected path. The total effective resistance R_{eff} is determined by the parallel connection of $R_M(T)$ and $R_I(T)$. Rapid change of f near T_c induces the sharp peak in the resistivity of the manganites.

In our simulation, the distribution of the FM sites or the PM sites with the temperature evolution is implemented by the Monte Carlo method. Taking an example for a cooling process, there are PM sites in the 100×100 matrix except for the definite FM cluster seeds first, which are randomly distributed, for the consequent FM clusters to grow around. Then, with the temperature decreasing, the FM fraction increases and the PM fraction varies oppositely. To represent this process, we alter some original PM sites to FM sites, where the number of the altered sites is determined by the increment of the FM site fraction. We confine that the FM sites may only be added around the existing clusters, which means that the correlation effect among the FM or PM sites is included. All are opposite in the warming process.

The $La_{2/3}Ca_{1/3}MnO_3$ film is grown on the $SrTiO_3$ substrate tilted by 10° towards the [010] direction. Figure 1 shows the variation of the resistivity of the film during cooling and warming processes, where the asterisks denote the experimental data observed in the cooling process and the circles denote the experimental data in the warming process. The simulated results are represented by the solid line for the cooling and by the dot-dot-dashed line for the warming. In the cooling from room temperature, the resistivity gradually increases and reaches a maximum at about 270 K. This is followed by a steep resistivity drop at the temperature of about 240 K, and then changes to 5 K slowly. In

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** Email: kjjin@aphy.iphy.ac.cn

the warming, the resistivity follows the cooling curve first, and then deviates from the knee temperature of the cooling process. After an abrupt lifting beginning at the temperature of about 250 K, the resistivity reaches a maximum at about 280 K, and finally coincides with the cooling curve at high temperature. This is because the fraction of FM regions varies differently in the cooling and warming processes, which has been mentioned in a previous report.[7] Figure 2 simulates the varying process of the FM/PM regions every 20 K in the warming procedure from 240 K to 300 K. The white regions denote the PM regions and the black regions denote the FM regions. It is clear that many PM clusters are dispersed in connected FM regions at 240 K. When the temperature increases, the PM clusters grow larger and connect at about 280 K and the FM regions are cut as many areas, leading to the MIT mentioned above.

To study the influence on the resistivity by external magnetic field, we carried out the above-mentioned experiments under an external magnetic field of 1 T. The results are presented in Figure 3. Figure 4 displays the relation between magnetoresistivity (MR, defined as $\frac{\rho_H(T)-\rho_0(T)}{\rho_0(T)}$) and temperature during

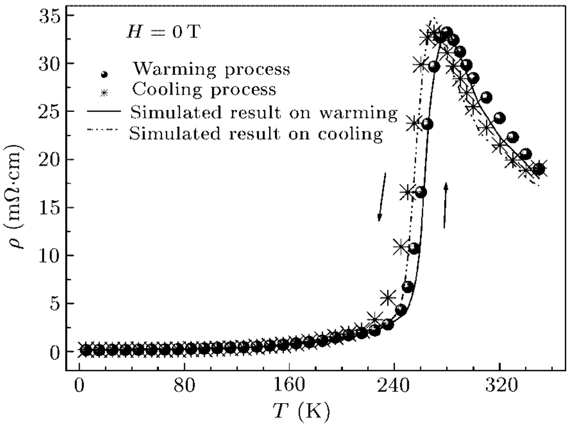

Fig. 1. Thermal hysteresis in resisitivity of $La_{2/3}Ca_{1/3}MnO_3$ thin films without external magnetic field. The arrows denote the direction of variation of the temperature.

Table 1. Parameters used in the simulation for $La_{2/3}Ca_{1/3}MnO_3$ thin films without and with H, respectively.

H (T)	ρ_{m0} (Ω·cm)	ρ_{m1} (Ω·cm·K^{-2})	ρ_{m2} (Ω·cm·K^{-4})	ρ_{i0} (Ω·cm)	E_0/k_B (K)
0	0.19	9.45×10^{-6}	2.62×10^{-11}	9.0	110
1	0.19	9.45×10^{-6}	2.36×10^{-11}	9.0	80

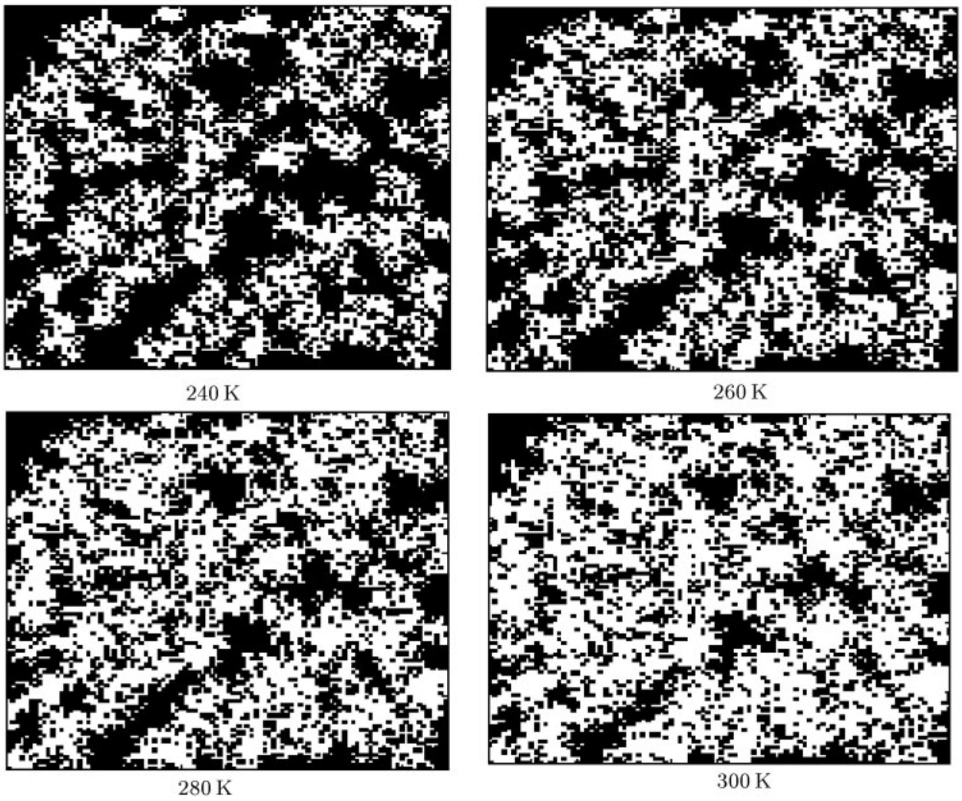

Fig. 2. The variation of the FM/PM regions during the warming process. The black regions denote the FM regions and the white regions denote the PM regions.

the warming and cooling processes, where the denotations of the symbols and lines are same as those in Figure 1. It is observed that the $\rho-T$ and MR-T curves also represent thermal hysteresis, indicating that the fraction of FM regions also varies differently in the cooling and warming processes under the external magnetic field. Furthermore, the temperature of MIT shifts to a higher value and the resistivity of the film is reduced in all the temperature ranges, that is, the films have negative MR. From the simulation parameters listed in Table 1, we find that only ρ_{m2} and E_0 are reduced by the external magnetic field but the other parameters are unchanged. The magnetic field can reduce the activation energy E_0, that is, make the PM insulating site in the film easier to become FM metallic site. It can be concluded that FM regions may be large enough to connect at a higher temperature so that the temperature of MIT increases by applying an external magnetic field. Meanwhile, the external magnetic field reduces the coefficient ρ_{m2} but barely influences the coefficients ρ_{m0} and ρ_{m1}, because the magnetic field reduces the spin scattering in conduction carriers by driving the local orientation of magnetization aligned. The above discussion indicates that our conclusion is reasonable.

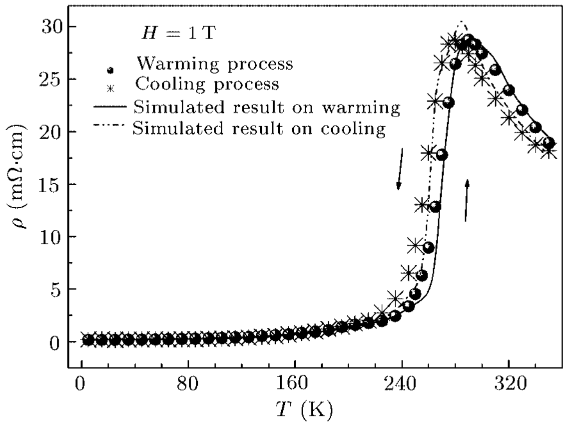

Fig. 3. Thermal hysteresis in resisitivity $La_{2/3}Ca_{1/3}MnO_3$ thin films with an external magnetic field of 1 T.

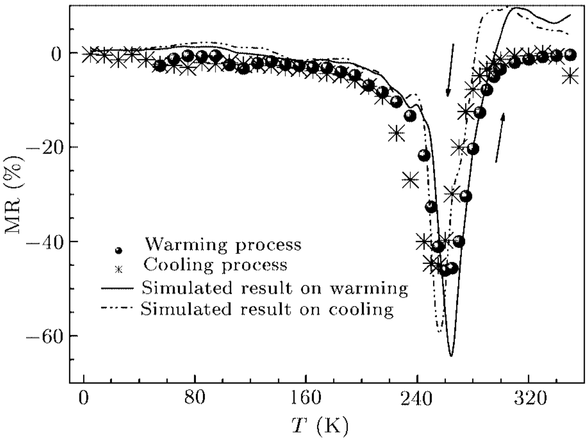

Fig. 4. Magnetoresistivities of $La_{2/3}Ca_{1/3}MnO_3$ thin films during the warming and cooling processes.

In summary, by using the random network model based on the phase separation scenario, we simulate the resistivity of $La_{2/3}Ca_{1/3}MnO_3$ thin films grown on tilted $SrTiO_3$ substrates with and without an external magnetic field of 1 T. The resistivities exhibit thermal hysteresis with and without the magnetic field, suggesting that the fraction of FM domains varies differently in cooling and warming processes and leading to the thermal hysteresis in the curves of MR versus temperature. The simulated results are in good agreement with the experimental data and suggest that the external magnetic field can increase the metallic-insulating transition temperature and can reduce the film resistivity.

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