Volume 9, Number 8
1009-1963/2000/09(08)/0630-04
CHINESE PHYSICS
August, 2000
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ENHANCEMENT OF FERROMAGNETIC CLUSTER INDUCED BY MAGNETIC FIELD IN THE PHASE-SEPARATED $La_{0.5}Ca_{0.5}MnO_3$*

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(Received 13 March 2000)

Magnetic and transport properties of $La_{0.5}Ca_{0.5}MnO_3$ have been investigated by measuring the magnetization and resistance in zero-field-cooled (ZFC) and field-cooled (FC) modes. Conspicuously irreversible behaviors of magnetization/resistance in the two different modes were observed below the charge ordering transition temperature (T_{CO}). The ZFC and FC magnetizations at 5K, as functions of the magnetic field, coincide for $\mu_0 H \leq 1$T. Afterwards, the ZFC magnetization tends to an approximate constant, but the FC one increases linearly with increasing field. There exists an excellent correspondence between magnetization and resistance below T_{CO}. All the results suggest that the ferromagnetic clusters embed in the charge-ordered matrix. The phenomenon of ferromagnetic clusters growing up easily in the FC procedure has been interpreted according to the model of thermally activated two-level system.

Keywords: ferromagnetic cluster, phase separation
PACC: 7560N, 7530K

I. INTRODUCTION

Rare earth manganites $Ln_{1-x}A_xMnO_3$ (Ln = rare earth, A = alkaline earth) have long been a subject of intensive study due to their interesting behaviors such as colossal magnetoresistance,[1−4] metal-insulator transition, and charge ordering (CO).[5−8] Double exchange mechanism proposed by Zener[9] provides a qualitative explanation for the magnetoresistance properties of this kind of materials. Afterwards, Millis *et al.*[10] proposed that the double exchange model alone cannot explain these complex magnetic and transport behaviors and the electron-lattice interaction has to be considered. However, quite recently, these materials have been considered as an inhomogeneous system in order to understand these complicated behaviors. Moreo *et al.*[11] proposed the phase separation scenario for perovskite manganite. Uehara *et al.*[12], from the viewpoint of percolative phase separation, interpreted why the magnetoresistive response increases dramatically when the Curie temperature (T_C) is reduced. By using scanning tunneling spectroscopy, Fäth *et al.*[13] observed the phase separation below T_C where inhomogeneous structures of metal and insulator areas coexist and are strongly field dependent in their size and structure in $La_{0.7}Ca_{0.3}MnO_3$ thin films. Based on the electronic microscopy observation, Mori *et al.*[14] found the coexistence of ferromagnetic and charge-ordered antiferromagnetic microdomains with the size of 20∼30nm in $La_{0.5}Ca_{0.5}MnO_3$. In this paper, we investigate the magnetic and transport properties of $La_{0.5}Ca_{0.5}MnO_3$ in zero field-cooled (ZFC) and field-cooled (FC) modes. All measurement results of magnetization and resistance suggest the existence of ferromagnetic (FM) clusters in the charge-ordered matrix, and the size of the FM cluster can increase dramatically in the FC procedure.

II. EXPERIMENTAL

The polycrystalline sample of $La_{0.5}Ca_{0.5}MnO_3$ was prepared by the conventional ceramic route. The well-mixed powders of La_2O_3, $CaCO_3$, and $MnCO_3$, with appropriate proportions were calcinated at 800°C in air for 24h. The resulting mixture was reground, pressed into pellets and sintered at 1300°C for 48h, then furnace-cooled to room temperature. X-ray diffraction, carried out using a Rigaku X-ray diffractometer with a rotating anode and Cu K_α radiation, indicates that the sample is a cubic single phase with the lattice parameter a=3.8354Å. Magnetization measurements were performed on an extracting sample magnetometer. Resistance was measured by the stan-

*Project supported by the National Natural Science Foundation of China and State Key Project of Fundamental Research of China (Grant No. G1998061305)

dard four-probe method with the current parallel to the magnetic field .

Ⅲ. RESULTS AND DISCUSSION

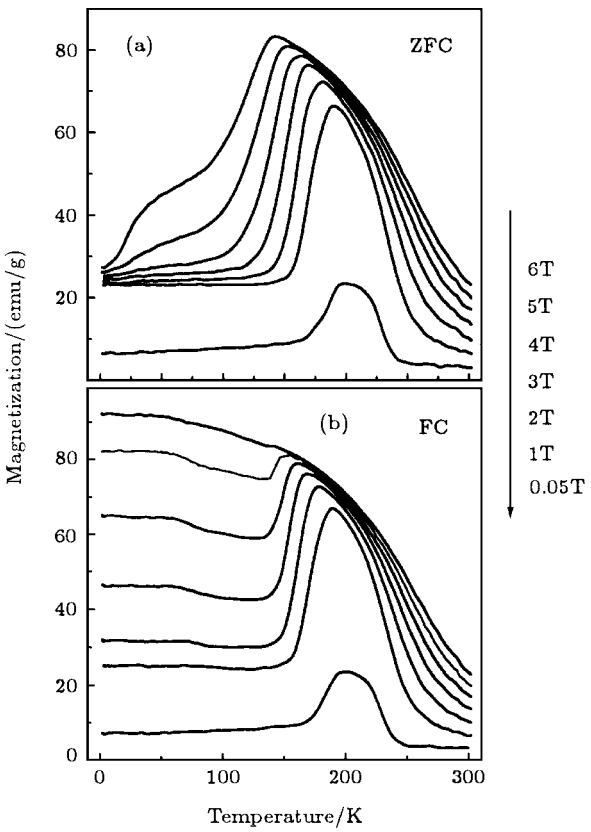


Fig.1. The temperature dependence of magnetization ($M-T$) measured in warming process under various fields (0.05, 1, 2, 3, 4, 5, and 6T) in ZFC mode (a) and FC mode (b).

Figure 1 shows the temperature dependence of magnetization ($M-T$) under various magnetic fields in ZFC and FC modes, respectively. When $\mu_0 H$=0.05T, the sample exhibits a paramagnetic to ferromagnetic transition at 245K followed by a ferromagnetic metal to antiferromagnetic (AFM) charge-ordering transition around 175K. It is noted that the magnetization (6.6emu/g) below the charge-ordering transition temperature ($T_{\rm CO}$) is much larger than that expected for an AFM phase. The FM ingredient in the sample should be responsible for the higher low-temperature magnetization. In both ZFC and FC processes, the $T_{\rm CO}$ reduces gradually with increasing field. In ZFC mode, the magnetization at 5K hardly varies with increasing field for $\mu_0 H \geq$1T, while the $M-T$ curve exhibits an obvious shoulder at about 50K when $\mu_0 H \geq$5T. However, a striking feature is that the low-temperature magnetization increases with increasing magnetic field in FC mode. In other words, the FC procedure enhances the FM ingredient. Finally, the charge-ordering transition disappears and the sample behaves as a typical ferromagnet. Interestingly, a step of the magnetization is induced in the FC process around 75K, which may be related to the undercooled FM phase in the FC sample.[15] Obviously, under a fixed field, the $M-T$ curves in ZFC and FC modes cannot coincide with each other below $T_{\rm CO}$. A higher magnetic field can magnify the irreversibility.

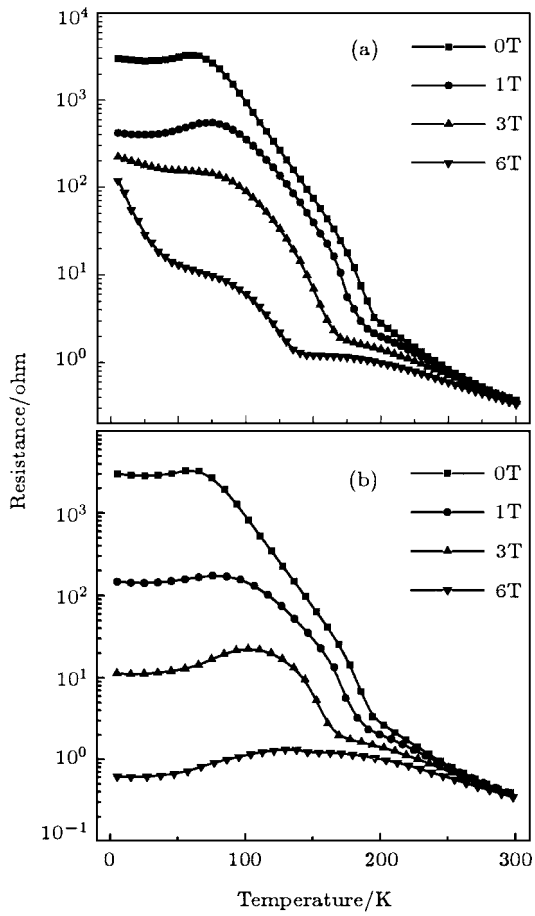


Fig.2. The temperature dependence of resistance measured in fields of 0T, 1T, 3T, and 6T, in ZFC mode (a) and FC mode (b).

Figure 2 presents the resistance as a function of temperature (R-T) under various fields in ZFC and in FC modes, respectively. When $\mu_0 H$=0, a weak metal-insulator transition can be observed at about 60K. Corresponding to the charge-ordering transition, a jump in resistance is obviously discernible around $T_{\rm CO}$. Under the same field, there is an excellent correspondence between the magnetization and resistance below $T_{\rm CO}$, the increase of magnetization corresponding to the decrease of resistance. The excellent correspondence is especially striking in ZFC mode un-

der a high field, as in the case of $\mu_0 H$=6T. The increase of magnetic field can change the temperature dependence of resistance in the ZFC sample, as shown in Fig.2(a). When $\mu_0 H$=6T, the sample behaves as an insulator in ZFC mode, but it exhibits a metal-insulator transition at 150K in FC mode. Similar to the magnetization, the ZFC and FC resistances are also irreversible. The irreversibility can also be magnified by a magnetic field. According to the equivalence between magnetization and resistance, one can conclude that the changes in resistance below T_{CO} may result mainly from spin-dependent scattering.

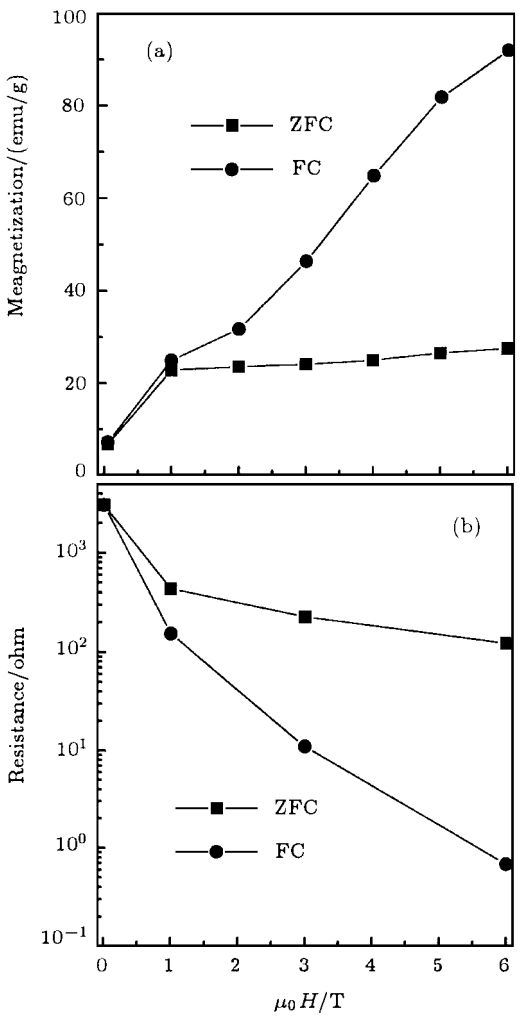


Fig.3. The magnetic field dependence of magnetization (a) and resistance (b) in ZFC and FC modes at 5K.

Figure 3 shows the magnetization and resistance as functions of field at 5K measured in ZFC and in FC modes, respectively. When $\mu_0 H \leq$1T, the ZFC and FC magnetizations coincide with each other. With further increase of field, the ZFC magnetization tends to an approximate constant, but the FC one increases linearly in spite of a singular point at $\mu_0 H$=1T. According to the phase diagram reported by Millis,[16] $La_{1-x}Ca_xMnO_3$ is of a FM phase in the samples for $0.2< x <0.5$ and charge-ordered AFM phase for $x > 0.5$. For the compound $La_{0.5}Ca_{0.5}MnO_3$, the higher low- temperature magnetization (Fig.1(a)) and the metal-insulator transition at a low temperature (Fig.2) both suggest the appearance of the FM phase due to the inhomogeneous oxygen distribution. From these irreversible behaviors of magnetization and resistance in ZFC and FC modes, it can be concluded that the FM clusters (oxygen-poor region) are embedded in the CO matrix (oxygen-rich region). The relative proportions of FM and CO phases can be evaluated as follows. The average moment of Mn ion (μ_{exp}) under a field of 1T, which is large enough for the domain rotation of FM clusters, and the saturation moment (μ_s) are $0.79\mu_B$ and $3.5\mu_B$, respectively. If the smaller magnetic moment of CO phase is considered to be negligible compared with the larger one of FM phase, the ratio of the two phases FM:CO approaches 23:77, based on the ratio of μ_{exp}/μ_s. In fact, there is a little amount of CO phase in the FC sample for $\mu_0 H$=6T in spite of its obvious FM behaviors. Similar evaluation indicates that the fraction of FM phase is increased up to 96% under a field of 6T in FC mode. It is thus clear that the FM clusters can grow up at the expense of the CO phase.

We can interpret the above results in terms of the model of thermally activated two-level system, as previously proposed for $Nd_{0.5}Sr_{0.5}MnO_3$ system.[5] In this model each of the FM metal phase and AFM insulator phase occupies a minimum in an asymmetrical well, while the lowest minimum being here occupied by the insulating state. As is well known, magnetic field can reduce the barrier and the FM state potential well. In the ZFC mode, the magnetic field cannot drive the system to climb up the barrier and crossover from CO state to the FM state at 5K. However, the ZFC magnetization tends to a constant for $\mu_0 H \geq$1T due to the domain rotation of all the FM clusters. Under a higher magnetic field ($\geq$5T), part of CO phase can melt into FM state due to the increase of temperature, which result in the magnetization shoulder at low temperature (Fig.1). In FC mode, the magnetic field favors all Mn moment, including the oxygen-poor and oxygen-rich regions, to align in the same direction, and FM clusters can grow up due to the aid of the magnetic field. On the other hand, the FM clusters can also pin the Mn moment around them ferromagnetically. Consequently, the undercooled FM state may remain

down to low temperatures. The low-temperature FM state induced by the FC process should be metastabe as is in the $(Nd,Sm)_{0.5}Sr_{0.5}MnO_3$ system reported by Tokura *et al.*[17]

IV. CONCLUSION

By measuring the magnetic and transport properties of $La_{0.5}Ca_{0.5}MnO_3$ in ZFC and FC modes, we found the large irreversibility both in magnetization and resistance below T_{CO}. The irreversibility can be magnified by the magnetic field. All the results support the conclusion that the FM clusters are embedded in the CO matrix. The FC procedure has more ability to promote the FM clusters to grow up.

ACKNOWLEDGMENTS

Thanks will go to T. S. Ning and S. Y. Fan for their help in measurements.

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