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The coexistence and competition of low-field magnetoresistance and colossal magnetoresistance in polycrystalline $La_{0.49}Sr_{0.51}(Mn_{1-x}Nb_x)O_3$

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Abstract

The magnetic and transport properties of polycrystalline $La_{0.49}Sr_{0.51}(Mn_{1-x}Nb_x)O_3$ have been studied by the measurement of dc magnetization and magnetoresistance (MR). With the increase of Nb doping, the ferromagnetism evolves from the inferior position in the $x=0$ sample to the dominant position in the $x=0.05$ and 0.15 samples, and then to the inferior position again in the $x=0.25$ sample. Therefore, there are not enough ferromagnetic domains to form percolative conduction paths in the $x=0$ and 0.25 samples, as a result, no obvious MR has been found under a 4500 Oe magnetic field in the two samples. But the intrinsic colossal MR (CMR) effect and the extrinsic low-field MR (LFMR) effect have been observed coexisting in the samples of $x=0.05$ and 0.15 under 4500 Oe. On the other hand, the anisotropic MR has been observed in the $x=0.05$ and 0.15 samples when a 4500 Oe field is respectively applied perpendicular and parallel to the plane of the pellet-shape samples. The anisotropic MR is attributed to the existence of LFMR.

Keywords: Magnetic materials; Magnetometer; Electrical characterization; Transport properties

1. Introduction

The effect of "colossal magnetoresistance (CMR)" in hole-doped manganites has attracted significant interest from researchers in the past decade [1–4] because of their rich physical properties and potential device applications. Their rich properties are due to the coupling and competition among interactions involving spin, lattice, and charge degrees of freedom. The underlying CMR mechanism is not yet fully understood, but phase separation (PS), where metallic ferromagnetic (FM) domains and insulating antiferromagnetic (AFM) regions coexist, is very important [5–7]. Besides their intrinsic CMR effect, polycrystalline ceramics of manganites exhibit extrinsic low-field magnetoresistance (LFMR) effect [7–10], which is more promising in applications. The latter effect is generally attributed to the spin-dependent scattering near grain boundaries [9] and/or the spin-polarized intergrain tunneling [8]. Recently, the competition and coexistence of CMR and LFMR in manganites are proved in theory by Ju et al. [7].

In this paper, we focus on the hole doping in polycrystalline $La_{0.49}Sr_{0.51}MnO_3$, which was achieved by the doping of Mn sites with the Nb^{5+} cations. In the neighborhood of $x=0.5$, polycrystalline $La_{1-x}Sr_xMnO_3$ is a PS system so that instead of having a uniform and homogenous magnetic ground state, the low-temperature phase is an intimate mixtures of FM, AFM and charge ordering (CO) regions in the grains of the ceramic sample [11]. Due to the existence of CO in $La_{0.49}Sr_{0.51}MnO_3$, the Nb^{5+} doping for Mn sites should first destroy the CO and induce the enhancement of FM as well as the insulator–metal transition according to Ref. [12], leading to rich physical properties. The magnetic and transport properties of the samples with different doping level as well as their MR behavior were

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carefully studied. Not only CMR but also LFMR were observed in polycrystalline $La_{0.49}Sr_{0.51}(Mn_{1-x}Nb_x)O_3$, and there exists competition between them.

2. Experimental

Polycrystalline samples of $La_{0.49}Sr_{0.51}(Mn_{1-x}Nb_x)O_3$ ($x = 0$, 0.05, 0.15 and 0.25) were prepared via the standard solid-state reaction route starting from high purity La_2O_3, $SrCO_3$, MnO_2 and Nb_2O_5 according to the experimental procedure previously reported [13]. X-ray diffraction data show that the prepared samples are single phase without any other secondary or impurity phase. The physical properties of the samples have been checked by means of dc magnetization (M) and four-probe resistivity (ρ). The temperature dependence of M was measured with a Foner vibrating sample magnetometer, the ρ measurement for zero field and 4500 Oe field was performed from 310 K down to 10 K within a Janis CSS300-EB closed cycle refrigerator system. It should be mentioned that all our samples were cut into pellets. During the measurement of MR, a magnetic field of 4500 Oe was respectively applied perpendicular to the pellet plane ($H_\perp$) and parallel to the pellet plane ($H_{||}$) in order to analyze the MR anisotropy.

3. Results and discussions

Fig. 1 shows the zero-field cooled (ZFC) M–T curves for the different doping level samples in a magnetic field of $H = 12$ kOe. The $x = 0$ sample first exhibits a transition from paramagnetic (PM) to FM with Curie temperature $T_C \approx 275$ K. With the decrease of temperature, M reaches a maximum value at about 170 K. After that, M shows a sharp decline with temperature decrease indicative of the transition from FM to AFM ($T_N \approx 115$ K) [11,14]. At low temperature, however, there is still a sizable nonzero M indicating that well below T_N, a considerable FM component exists in the material. Therefore, $La_{0.49}Sr_{0.51}MnO_3$ is a PS system with the coexistence and competition of several phases, i.e. FM and AFM states, even CO state according to Ref. [11]. Compared to the $x = 0$ sample, the PM-FM transition still exists for all doping samples, but the FM-AFM transition disappears. The Curie temperature first increases a little with the increase of doping level, and then decreases rapidly to lower temperature, i.e. $T_C \approx 280$ K, 104 K and 100 K for the $x = 0.05$, 0.15 and 0.25 samples, respectively. As far as the M values at low temperature are concerned, the same evolution is found with the doping level, i.e. M first rises and then

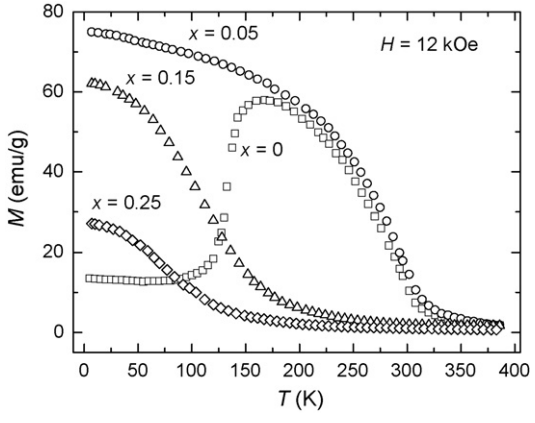

Fig. 1. Temperature dependence of the magnetization of $La_{0.49}Sr_{0.51}(Mn_{1-x}Nb_x)O_3$ for ZFC under a 12 kOe field.

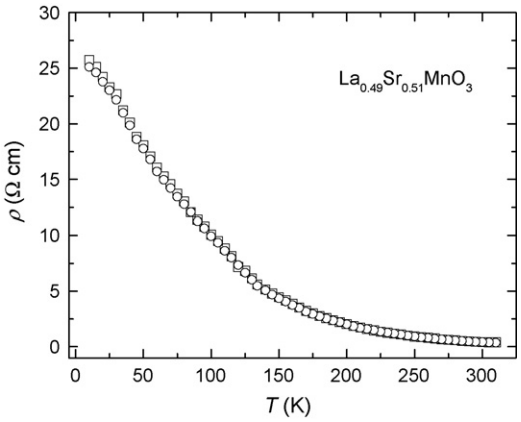

Fig. 2. The resistivity vs. temperature of $La_{0.49}Sr_{0.51}MnO_3$ for zero field (square) and 4500 Oe magnetic field (circle). The field is applied parallel ($H_{||}$) to the plane of the pellet-shape sample.

declines with the increase of Nb content. The above phenomena can be interpreted as follows, the introduction of Nb^{5+} cations into Mn sites can induce two effects: (1) destroy the CO state in $La_{0.49}Sr_{0.51}MnO_3$ and result in the enhancement of FM; (2) the diamagnetic Nb^{5+} cations do not participate in exchange interactions and destroy the double exchange between Mn^{3+} and Mn^{4+} [12]. When the Nb content is low, the former dominates the evolution of T_C and M value. However, with the further increase of doping level, the influence of the latter can prevail over that of the former.

The ρ–T curves of the $x = 0$ sample for zero field as well as for 4500 Oe field were measured in the parallel-field geometry and shown in Fig. 2. It is found that there is almost no change for the resistivity under a magnetic field of 4500 Oe compared to that of zero field. On the other hand, no insulator–metal transition corresponding to the PM-FM transition in M–T curve happens for the ρ–T results whether the magnetic field is applied or not. From the M–T measurement, it is known that there coexists FM, AFM and CO phases in $La_{0.49}Sr_{0.51}MnO_3$. In the PS manganites, the CMR effect can be explained by percolative transport through the FM domains, this depends sensitively on the relative spin orientation of adjacent FM domains which can be controlled by applied magnetic fields [5,6,15]. Despite the existence of FM component in the $x = 0$ sample, we suggest that no percolative conduction paths form between the FM domains, even if a 4500 Oe field is applied. Therefore, no insulator–metal transition and obvious MR can be found in the ρ–T curves for the $x = 0$ sample.

For the $x = 0.05$ sample, its ρ–T curves for zero field and for 4500 Oe field with two field directions ($H_\perp$ and $H_{||}$) are exhibited in Fig. 3. Fig. 3 also shows the corresponding MR ($\Delta\rho/\rho_H = [\rho_0 - \rho_H]/\rho_H$) ratios. We find that 5% Nb doping can induce the notable insulator–metal transition with resistivity peak ($T_P \approx 260$ K) near its Curie temperature ($T_C \approx 280$ K), which means the FM domains are sufficient to form percolative paths in this sample. It is consistent with the magnetization result, where the FM phase becomes dominant below T_C. It must be mentioned that our M–T and ρ–T measurements were performed in different magnetic field, the applied field is 12 kOe