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Simultaneous observation of positive and negative giant magnetoresistances in composite $(La_{0.83}Sr_{0.17}MnO_3)_{1-x}(ITO)_x$*

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We have studied the transport property of the composites $(La_{0.83}Sr_{0.17}\ MnO_3)_{1-x}(ITO)_x$ [ITO=$(In_2O_3)_{0.95}(SnO_2)_{0.05}$], which were fabricated by mechanically mixing $La_{0.83}Sr_{0.17}MnO_3$ and ITO grains. A giant positive magnetoresistance (PMR) has been observed above the Curie temperature T_c for samples with x around 0.40, in addition to the negative magnetoresistance related to spin-dependent interfacial tunnelling below T_c. For $(La_{0.83}Sr_{0.17}MnO_3)_{0.6}(ITO)_{0.4}$, the magnetoresistive ratio for the PMR can reach 39.3% under a magnetic field H=2.24×10^{5}A/m. Theoretical analysis suggests that the magnetic-field-induced broadening of the p–n barrier between both kinds of grains and the density of the p–n heterostructures should be responsible for the PMR behaviour.

Keywords: granular system, composite, giant magnetoresistance, p-n heterostructure
PACC: 7560J, 8160H, 7530V, 7340L

1. Introduction

Since 1990's, the hole-doped manganese oxides $La_{1-x}M_xMnO_3$ (M = Ca, Sr, and Ba etc.) have excited great interest due to their rich physical properties, such as colossal negative magnetoresistance (CMR), charge ordering, giant magnetostriction, and the large change of magnetic entropy etc.[1−4] The parent compound without divalent ions $LaMnO_3$ is an antiferromagnetic insulator. The divalent substitution leads to ferromagnetic order and metallic conductivity due to the double exchange (DE) between Mn^{3+} and Mn^{4+} ions.[5] For $La_{1-x}Sr_xMnO_3$, the Curie temperature $T_c$varies from 180 to 375 K when $0.12 < x < 0.45$. Under an applied field, T_c can be moved to a higher temperature, resulting in a CMR effect near T_c.

Later, a giant intergranular tunnel-type magnetoresistance (ITMR) has been observed in the perovskite granules.[6,7] The ITMR is also negative but can exist in the whole range of temperature below T_c and needs a smaller inducing magnetic field compared with the intrinsic CMR in the crystals. So it has more practical significance. The study on spin-dependent tunnelling or dispersion at particle boundaries related to ITMR attracted considerable attention after that.[8−10] Since the surface of the perovskite grains is in a semi-paramagnetic state due to the existence of a large number of dangling bonds, the interfaces between neighbouring grains could play a role of tunnelling barrier as compared with the kernels in the view of DE.[7]

Meanwhile, some attention has been paid to the composite systems of magnetic and nonmagnetic particles, which form some percolation systems. A larger enhancement of ITMR can be expected for the samples close to the percolation threshold.[11−14]

The study of giant positive magnetoresistance (PMR), which is of the same significant as giant negative one (NMR) in applications, has also stimulated considerable interest since Xu *et al* reported a PMR behaviour in non-magnetic silver chalcogenides, which are doped narrow-band semiconductors. The magnetoresistance ratio (MR) (defined as $(R_H - R_0)/R_0$, where R_H and R_0 are the resistances with and without

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a magnetic field respectively) for the PMR can reach 350% under a field H=4.4×10^{4}A/m at 4.5K. They considered that the effective mass of the electron in the material is smaller, so the classical orbital magnetoresistance can be greatly increased, resulting in a large PMR.[15] Subsequently, Zhang *et al* observed a PMR effect in $Nd_{0.67}Cs_{0.33}MnO_3$. The MR is as high as 10000% below the metal–insulator transition temperature (around 45K). They believe that the PMR originates from a field-induced carrier localization.[16] Manyala *et al* found an anomalous PMR in ferromagnet $Fe_{1-x}Co_xSi$. They argued that it arises by a quantum interference effect (QIE).[17] Chen *et al* reported another QIE-related PMR in $La_{0.7}Pb_{0.3}MnO_3$ single crystal between 4.2 and 50K and the MR is about 20% at T= 4.2 K under a magnetic field of $\mu_0 H$= 8T.[18] Recently, studies on some magnetic tunnel junctions and magnetic multilayer p–n heterostructures composed of hole-doped manganite perovskite have attracted more attention.[19–22] For example, a PMR has been discovered in a multilayer p–n heterostructures fabricated with Sr-doped $LaMnO_3$ and Nb-doped $SrTiO_3$ in the temperature range from 100 to 300 K and the MR can reach 517%.[21] Such PMR is regarded as an interfacial effect. Jin *et al* suggested, based on weak Hund's coupling, that the PMR is attributed to the different distributions of energy bands between both ends of the p–n heterostructure interface.[22]

Here, we present an observation of another kind of PMR in the composite $(La_{0.83}Sr_{0.17}\ MnO_3)_{0.6}[ITO]_{0.4}$ [ITO = $(In_2O_3)_{0.95}$ $(SnO_2)_{0.05}$] at higher temperatures simultaneous with the negative ITMR. $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) is a typical hole-doped manganese perovskite. Studies show that LSMO behaves like a ferromagnetic metal below T_c and changes into a p-type semiconductor above T_c.[23] While, indium tin oxide (ITO) is an n-type one with higher conductance, and often used to make conductive glass. When both are connected, a p–n heterostructure can form at the interface. We believe that the behaviour of the p–n barrier under an applied field and an appropriate density of the p–n junctions in the samples should be responsible for the giant PMR behaviour.

2. Sample preparation

The precursor powder LSMO used was synthesized by the sol-gel method as reported previously.[7] Then the LSMO powder was heated at 1000°C for 5h in air. ITO powder was obtained by ball-milling commercial ITO bulks. Both powders were mixed in terms of Wt % desired with ultrasonic vibration to obtain $(LSMO)_{1-x}(ITO)_x$ composite. The mixture was pressed into small strips and then sintered at 1250°C for 10h and cooled in furnace in an air atmosphere.

The structure of the samples was identified by x-ray diffraction (XRD). Their magnetization was measured by a vibrating sample magnetometer. Measurements of the resistance and the magnetoresistance were performed by using a standard four-probe technique and an applied field.

3. Results and discussion

The XRD spectra of the 1250°C sintered composites $(LSMO)_{1-x}(ITO)_x$ (x=0.0, 0.2 and 0.4) display two different sets of lines, which can be indexed by pure LSMO and ITO, respectively, as shown in Fig.1. We can see that the intensity of the lines indexed by ITO increases with increasing x, and no other peaks can be observed in the samples. That means no new phases have been produced in the mixing and subsequent sintering processes. In other words, no observable chemical reaction occurs between LSMO and ITO.

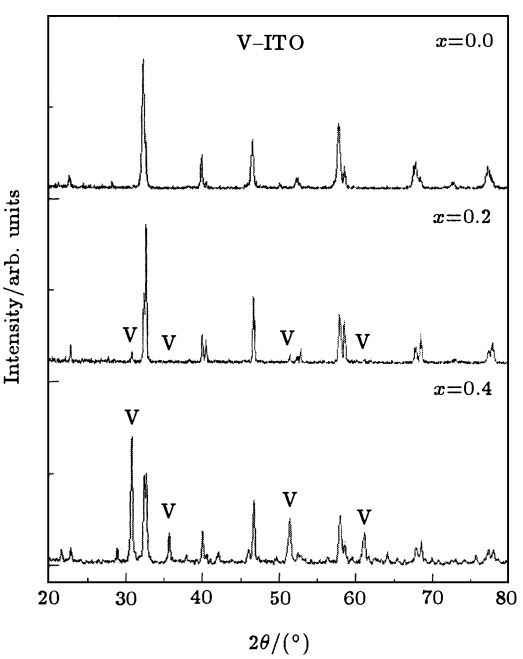


Fig.1. XRD spectra of composites $(LSMO)_{1-x}(ITO)_x$ (x=0.0, 0.2 and 0.4).

The temperature dependence of the magnetization for the 1250°C sintered $(LSMO)_{1-x}$ $(ITO)_x$ (x=0.0, 0.2 and 0.4) under an applied field $\mu_0 H$= 16000A/m is shown in Fig.2. As can be seen, their ferromagnetic transition temperatures T_c are almost the

same, at 287K, giving further evidence that no chemical reaction has taken place between LSMO and ITO in the view of magnetism. By the way, the magnetization of $(LSMO)_{0.8}(ITO)_{0.2}$ and $(LSMO)_{0.6}(ITO)_{0.4}$ reduces in turn due to the dilution effect of ITO.

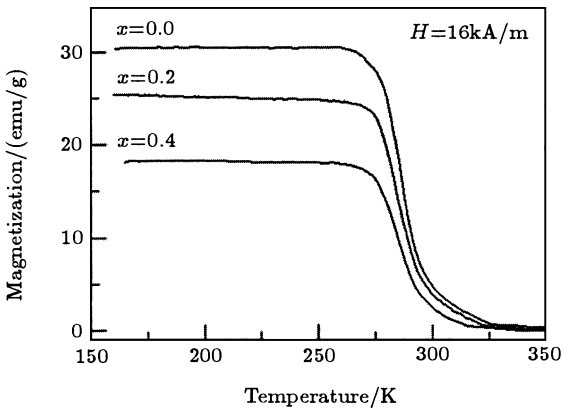


Fig.2. Temperature dependence of magnetization for composites $(LSMO)_{1-x}(ITO)_x$ (x=0.0, 0.2 and 0.4).

Figure 3 shows the temperature dependence of resistivity, ρ-T, without and with an applied field $\approx 2.24 \times 10^5$A/m, together with the corresponding magnetoresistance for the sample $(LSMO)_{0.6}(ITO)_{0.4}$. The MR defined here is the same as MR=$(\rho_H - \rho_0)/\rho_0$. It can be seen from the figure, a sharp resistivity peak appears at about 330K besides the small vale related to the intrinsic metal-insulator transition near T_c.

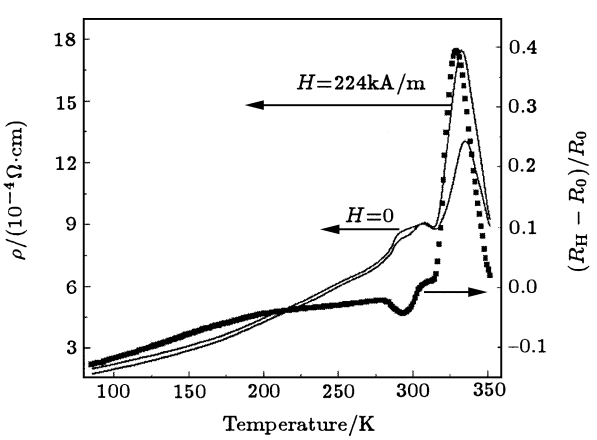


Fig.3. The temperature dependence of the resistivity without and with an applied field H=224kA/m, and the corresponding magnetoresistive ratio MR (the dot line) for the composite $(LSMO)_{0.60}(ITO)_{0.40}$.

Correspondingly, a giant PMR is observed between 320 and 345K. The MR for the PMR can reach as high as 39.3% under that field $\approx 2.24 \times 10^5$A/m. Meanwhile, an NMR has also been observed below T_c. Such NMR has the characteristic of interfacial tunnel-type magnetoresistance, that is, its absolute value decreases with increasing temperature.[11] Its MR can reach –13% also under the field $\approx 2.24 \times 10^5$A/m. It is much larger than that observed in pure La–Sr–Mn–O granules, considering that the applied field is so small.[11] This may be related to a percolation effect of the composite system.[12]

The origination of the PMR observed here seems not to be the same as that mentioned previously. ITO is an n-type semiconductor and LSMO is a p-type one above T_c. After the mixing of both constituents, p–n heterostructures can form everywhere in the sample. Thus, the transport property of the sample will closely related to these p–n heterostructures. It is well known that a space charge region and built-in electric field can form at both ends of a p–n junction as a result of the difference between the carrier density gradients in both zones. The built-in field plays a role of potential barrier for carriers. Thus the width of the barrier determines the resistance of the p–n junction, consequently, determines the conductance of the sample. The resistivity ρ of the p–n junction can approximately be expressed as

$$\rho \propto \frac{1}{V} e^{\frac{KV}{D}d}, \tag{1}$$

where V is the diffusion velocity, which is proportional to the transfer velocity of carriers, and the transfer velocity is further proportional to $T^{-3/2}$. D is the diffusion coefficient, which is, according to Einstein relation, proportional to $T^{-1/2}$. K is a constant, and d is the width of the barrier. Consequently, Eq.(1) can be rewritten as

$$\rho = k_1 T^{\frac{3}{2}} e^{k_2 d/T}, \tag{2}$$

where k_1 and k_2 are two constants also.

The resistance of LSMO grain tends to reduce when it is in a magnetic field. That is to say, the hole concentration in the p-zone increases. As a result, the carriers in the p–n heterostructures can diffuse further, and result in the barrier widening. Accordingly, the resistance of the sample gets higher. That leads to a PMR. The sharp resistivity peak near 330 K can also be understood from Eq.(2). Additionally, we have noticed that the PMR is only observed in the samples with x in a narrow range around 0.40 and the MR rapidly decreases when x deviates 0.40, as shown in Fig.4. We guess that the appearance of the PMR may rely on the density of p–n heterostructures in the samples. Thus we believe that 0.40 is some kind of threshold for the PMR observed in the system.

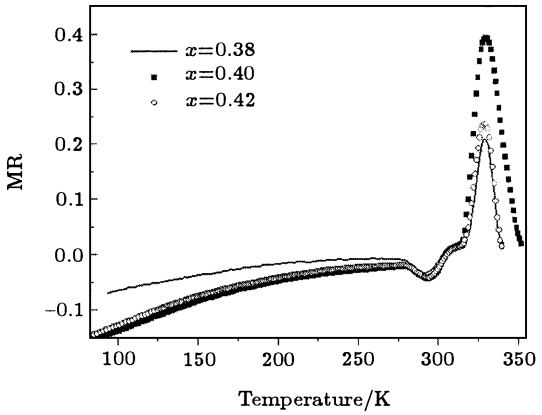


Fig.4. The temperature dependence of the MR for the composites $(LSMO)_{0.62}(ITO)_{0.38}$, $(LSMO)_{0.60}(ITO)_{0.40}$ and $(LSMO)_{0.58}(ITO)_{0.42}$.

4. Conclusion

A giant positive magnetoresistance has been observed in composite $(La_{0.83}Sr_{0.17}MnO_3)_{0.6}(ITO)_{0.4}$ between 320K and 345K. The MR is as high as 39.3% under a magnetic field 224kA/m. Theoretical analysis shows that the magnetic-field-induced broadening of the p–n barrier between the grains of LSMO and ITO and an appropriate density of the p–n heterostructures in the sample should be responsible for the positive magnetoresistance behaviour.

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