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The effect of MgO doping on the structural, magnetic, and magnetotransport properties of $La_{0.8}Sr_{0.2}MnO_3$ manganite

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Abstract

The effect of MgO doping on the structural, magnetic, and magnetotransport properties of $La_{0.8}Sr_{0.2}MnO_3$ (LSMO)/*x*MgO has been investigated. All samples were prepared by a solid-state reaction method. Alternating current susceptibility measurements for LSMO/*x*MgO samples show that the Curie transition temperature (T_c) and magnetization decrease with the increase of MgO concentration. The rate of the decrease of T_c at higher doping level is very fast. Also, samples with low doping level ($x \leq 2$) show insulator-metal transition, but the transition temperature decreases and resistivity increases with the amount of MgO. It is observed that there is no insulator-metal transition at higher MgO doping level ($x \geq 3$). Also, the results show that the value of low-field magnetoresistance decreases with the increase of MgO doping level. It seems that due to the higher sintering temperature and almost the same ionic radii of Mg^{2+} and Mn^{3+}, Mg^{2+} mostly replaced Mn^{3+} and weakened the double-exchange interaction, and consequently, T_c and magnetoresistance decrease and resistivity increases with MgO doping.

Keywords: Manganites, Magnetoresistance, MgO, Doping

PACS: 71.30.Th, 75.47.Lx, 75.47.Gk

Introduction

Perovskite manganite with the formula $La_{1-x}A_xMnO_3$ (A = Sr, Ca, Ba, or vacancies) has attracted considerable attention due to the discovery of the phenomenon of colossal magnetoresistance (CMR) and its potential application [1-3]. The properties of this material are explained by the double-exchange theory of Zener [4] and electron lattice interaction [5]. However, the intrinsic CMR effect in perovskite manganites, which is caused by the double-exchange mechanism, is found on a magnetic field scale of several teslas and in the narrow temperature range.

Recently, the effect of grain boundaries in perovskite manganites has been studied intensively [6-12]. It was found that the presence of grain boundaries in polycrystalline samples leads to a large low-field magnetoresistance (LFMR) effect over a wide temperature range below the Curie temperature (T_c). To achieve a LFMR, different extrinsic properties are manipulated. One of them is the grain size effect. In granular manganites, by decreasing the grain size, magnetoresistance increases, which is related to spin-dependent scattering and tunneling between neighboring grains [6]. Another way to increase LFMR is the mixing of these CMR materials with secondary phases. They include $La_{0.8}Sr_{0.2}MnO_3$ (LSMO)/TiO_2 [13], LCMO/Al_2O_3 [14], and LCMO/BN [15].

LFMR is generally thought to be extrinsic in nature and ascribed to spin-polarized tunneling of conduction electrons through grain boundaries as proposed by Hwang et al. [6]. The basic objective is to increase the height of the tunnel barrier between neighboring ferromagnetic grains. In this work, we choose MgO as the secondary phase to modify the grain boundaries. MgO is a very suitable candidate as a second-phase material because it is a nice insulator below room temperature.

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Experimental

The LSMO/*x*MgO samples, with $x = 0$, 1, 2, 3, 5, and 7.5 wt.%, were prepared by two steps. First, LSMO powders were prepared by a conventional solid-state reaction method. High-purity powders of La_2O_3, $SrCO_3$, and MnO_2 were mixed in stoichiometric proportions and sintered at 1,200°C for 24 h. The sintering and grinding procedures were repeated three times. In order to obtain very fine and homogenous samples, the resulting powders were subjected to high-energy ball milling using a planetary mill for 1 h. MgO with smaller sized particles were prepared by high-energy ball milling for 2 h. Second, appropriate amounts of LSMO and MgO powders were mixed, and homogeneous powders were formed. Finally, the mixtures were palletized at a pressure of 500 MPa and sintered at 1,250°C for 24 h.

The resistivity measurements were carried out by the four-probe method in a magnetic field of 5,000 Oe, using a Leybold closed-cycle refrigerator (Oerlikon, Cologne, Germany) at low temperatures down to 20 K. The alternating current (AC) susceptibility measurements were performed using a Lake Shore AC susceptometer (Model 7000, Westerville, OH, USA). X-ray diffraction (XRD) patterns of samples were taken on a Philips XPERT X-ray diffractometer. The microstructure of the samples was taken on a Philips XL 30 scanning electron microscope (SEM; Amsterdam, The Netherlands).

Results and discussion

Figure 1 shows the XRD patterns at room temperature for pure and doped samples. The XRD analyses reveal that all samples have a rhombohedral structure. As one can see, in lower concentrations of MgO doping, it is hard to see the peaks related to MgO and other impurities in composites, and LSMO lines are observed clearly. However, there is a small peak observed at $2\theta \approx 43°$ at high concentrations of MgO doping. This peak is related to MgO in the composites, and as can be seen, the peak intensity is very low. This may suggest that the MgO goes mainly to the perovskite structure and a very little part of the MgO goes to the grain boundaries. It seems that due to the higher sintering temperature, MgO has made a reaction with LSMO,

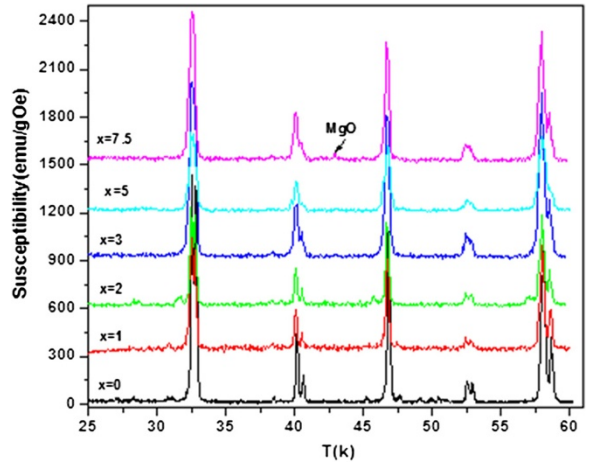

Figure 1 XRD patterns of LSMO/*x*MgO samples.

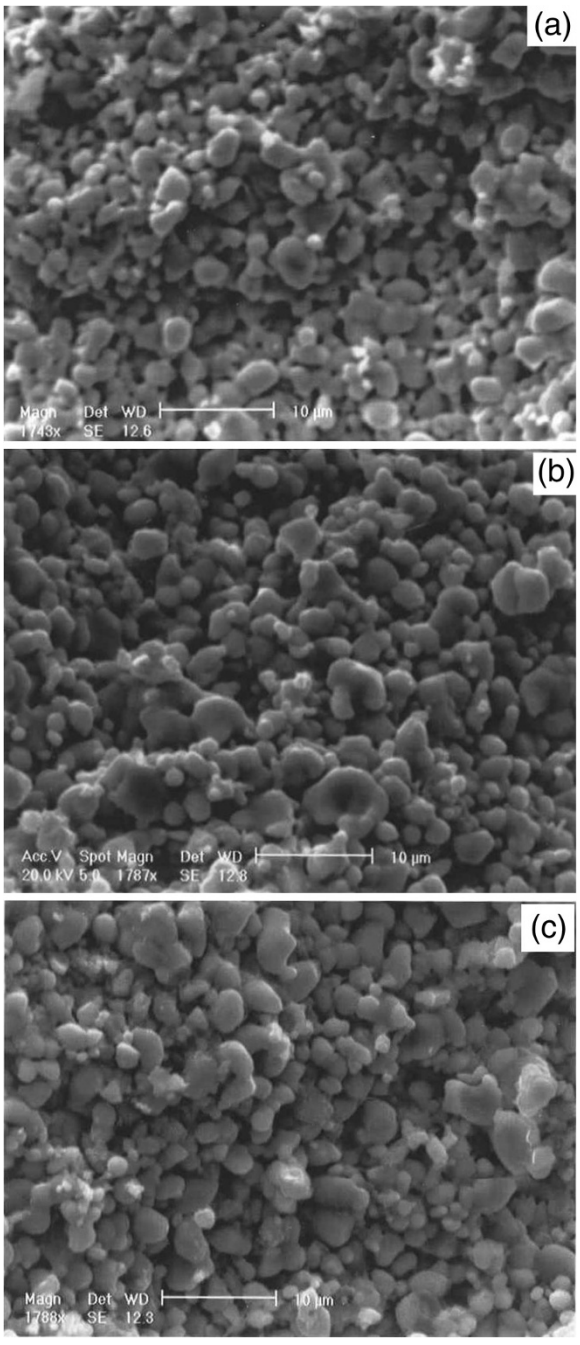

Figure 2 SEM micrographs of the samples. (**a**) $x = 0$, (**b**) $x = 3$, and (**c**) $x = 7.5$ wt.%.

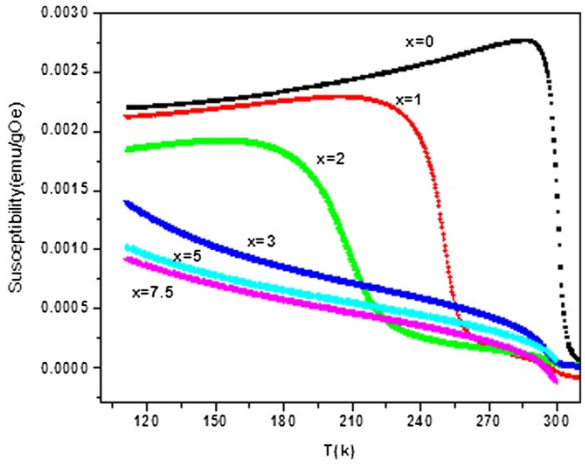

Figure 3 Temperature dependence of the real part of AC suceptibility for LSMO/xMgO samples in an AC field of 500 A/m at a frequency of 333Hz.

and Mg^{2+} mostly replaced Mn^{3+} into the perovskite structure. The same behavior was seen in the TiO_2-doped samples [13].

In order to analyze the effects of MgO on the microstructure of LSMO grains and the distribution of MgO in the composites, the fracture sections of all the samples were examined using the SEM. The typical SEM micrographs of the composites ($x = 0$, 3, and 7.5 wt.%) are shown in Figure 2. As shown, an almost clearer grain boundary is observed in all the samples, and there is no any trace of MgO even at high doping level. This means that most of the MgO goes into the perovskite lattice and probably a little part of the MgO is in the grain boundaries. This amount is very low, and it is not visible in the SEM micrographs.

Figure 3 shows the temperature dependence of the real part of AC susceptibility for the LSMO/*x*MgO samples in the AC field amplitude of 500 A/m at a frequency of 333 Hz. As one can see, the paramagnetic-ferromagnetic T_c and magnetization decrease with the increase of MgO concentration. Also, the rate of the decrease of T_c at higher doping level is very fast. These results show that the substitution of the Mg^{2+} ion for the Mn^{3+} ion weakens the double-exchange (DE) interaction significantly, and as a result, T_c and magnetization decrease because of magnetic dilution. These behaviors were confirmed by XRD analyses.

Figure 4 shows the resistivity versus temperature plot of the LSMO/*x*MgO samples in a zero magnetic field. As one can notice, the samples with low doping level ($x \leq 2$) show insulator-metal transition, but the transition temperature decreases and resistivity increases with the amount of MgO doping level. Also, there is no insulator-metal transition at high doping level ($x \geq 3$). These behaviors can be explained by the structural disorder [16]. As discussed in the

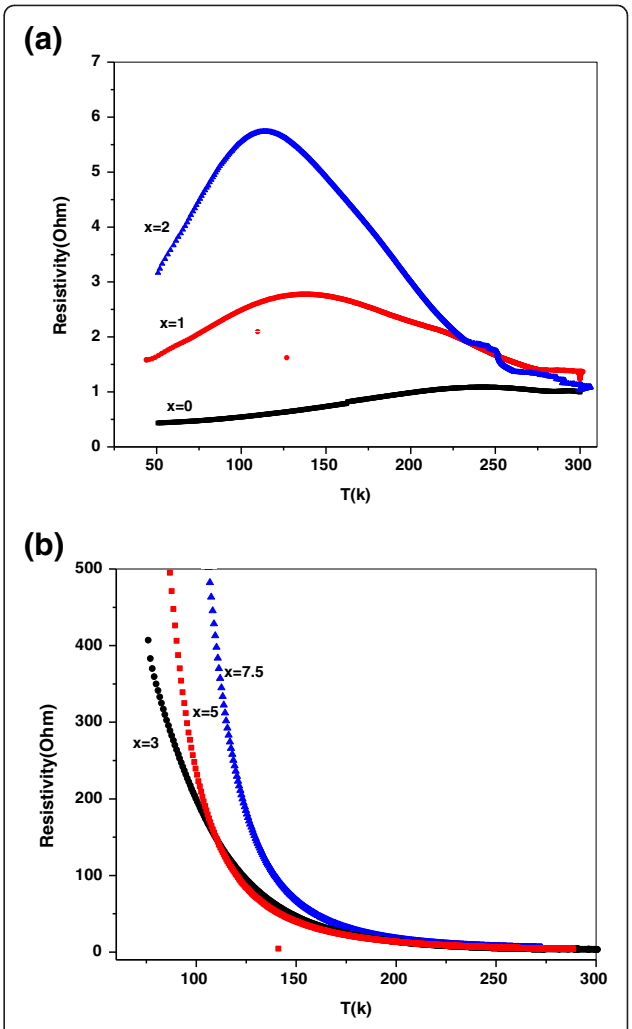

Figure 4 Resistivity versus temperature of LSMO/*x*MgO samples in a zero magnetic field. (**a**) $x = 0$, 1, 2 and (**b**) $x = 3$, 5, 7.5 wt.%.

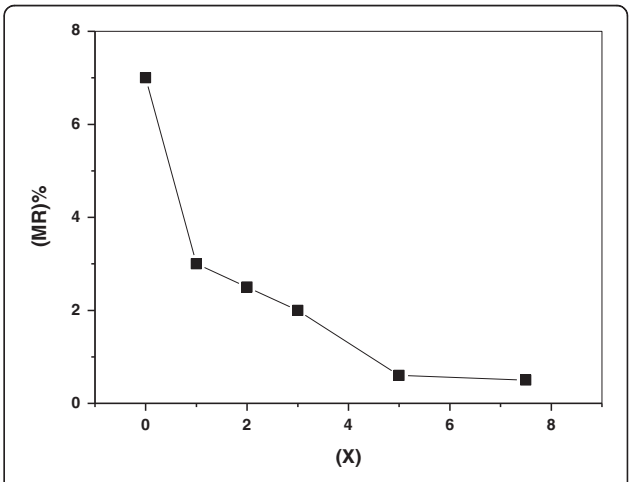

Figure 5 MgO doping (x) dependence of MR for LSMO/xMgO samples at a magnetic field of 5000 Oe and a temperature of 240 K.

literatures [17], there are two kinds of conduction channels, connected in parallel in the polycrystalline LSMO/xMgO composite samples. One is related to intragrain and the second is related to intergrain hopping of conduction electrons between neighboring sites. Moreover, the resistance of grain boundaries is more than the resistance inside the grains because of the disordered nature of the grain boundaries. With MgO doping at high concentrations, most part of the Mg goes inside the grains, substituting the Mn site in the LSMO lattice, and the remainder goes to the grain boundaries. If Mg substitutes Mn in the perovskite lattice, it cannot participate in the DE mechanism involving Mn^{3+} and Mn^{4+} ions, so, in effect, it dilutes the DE process. For that reason with higher doping, the DE mechanism decreases quickly, so resistivity and transition temperature increases and decreases very fast, respectively.

Figure 5 shows the LFMR for the LSMO/xMgO samples in a magnetic field of 5,000 Oe. It can be seen that the value of LFMR decreases with the increase of MgO doping level. As mentioned before, due to the large amount of substitution of Mg^{2+} for Mn^{3+}, the mobility of charge carriers reduces (since DE is weakened). This reduction of carrier concentration leads to an increase in resistivity and decrease in LFMR for all the samples. It seems that due to the higher sintering temperature and almost the same ionic radii of Mg^{2+} and Mn^{3+}, Mg^{2+} mostly replaced Mn^{3+} sites and weakened the double-exchange interaction, and consequently, T_c and magnetoresistance decrease and resistivity increases with MgO doping. On the other hand, the undoped sample has the maximum LFMR and transition temperature. Therefore, low-temperature sintering may be the next project to investigate the effect of MgO doping on LFMR in LSMO manganite samples.

Conclusions

The $La_{0.8}Sr_{0.2}MnO_3$/xMgO samples with $x = 0$, 1, 2, 3, 5, and 7.5 wt.% were prepared by a solid-state reaction method. The results of AC susceptibility measurements indicate that MgO doping decreases the Curie transition temperature and magnetization of the samples. Also, the results of resistivity measurements show that the resistivity increases and the insulator-metal transition temperature decreases with the increase of the MgO doping level. It is observed that there is no insulator-metal transition for the samples with a doping level of $x \geq 3$. Results also show that the value of low-field magnetoresistance decreases with the increase of MgO doping levels due to the higher sintering temperature and consequently replacement of Mg^{2+} in Mn^{3+} sites. On the other hand, the undoped sample has the maximum LFMR and transition temperature.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

ME participated in the preparation of the samples. PK participated in the characterization of the samples as well as in drafting the manuscript. HS participated in the analysis of the data and in editing the manuscript. All authors read and approved the final manuscript.

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