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The temperature dependence of the low field magnetoresistance in electron-doped manganites: $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$)

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Abstract

The low field magnetoresistance (LFMR) effect for an electron-doped manganese oxide: $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$) was investigated over the whole experimental temperature range of 5–300 K. A large LFMR of the samples was observed when the temperature was far above the Curie temperature and when it was far below the metal–insulator transition temperature. The experimental results—electron spin resonance spectra and the magnetic field dependence of the resistivity—suggested that the spin-polarized tunnelling between isolated ferromagnetic metallic clusters should be mainly responsible for generating the larger LFMR.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, perovskite manganites with a formula $R_{1-x}A_xMnO_3$ (R represents a rare earth and A is a divalent cation) have attracted considerable research interest due to the discovery of the colossal magnetoresistance (CMR) effect [1, 2] and the low field magnetoresistance (LFMR) effect [3, 4]. Generally, the CMR which originates mainly from the field-induced alignment of Mn spins is believed to be the intrinsic or intragrain magnetoresistance, which occurs in the vicinity of the Curie temperature (T_c) under a high magnetic field (several teslas). The LFMR is attributed as the extrinsic or intergrain magnetoresistance, which may stem from the spin-polarized tunnelling (SPT) [3, 5, 6] or spin-dependent scattering (SDS) [7] between neighbouring ferromagnetic grains. In particular, SPT was recently considered by Balcells *et al* [8] and Raychaudhuri *et al* [9] to be a predominant mechanism. No matter what the LFMR comes mainly from, the SPT or SDS, the grain boundaries play an essential role in generating the LFMR. Many experiments indicate that the significant LFMR of $R_{1-x}A_xMnO_3$

is only observed in polycrystalline samples including the forms of thin films, bulk ceramics, and ultrafine powders [10–13]. However, in most cases, the polycrystalline samples with large LFMR are hole-doped manganese oxides. Furthermore, the large LFMR appears far below T_c and rapidly decays with increasing temperature. In this paper, we report the low field magnetoresistance effect for $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$), which is an electron-doped manganese oxide with a colossal magnetoresistance effect [14]. The large LFMR of $La_{1-x}Te_xMnO_3$ was observed when the temperature was far above T_c or far below the transition temperature T_{MI} at which the metal–insulator transition occurred. In the temperature range of T_{MI} to T_c, there exist striking phase-separated features accompanied simultaneously by a CMR instead of a LFMR effect.

2. Experiments

Bulk $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$) was prepared by conventional ceramic techniques. The growth process and the phase structures have been described in detail elsewhere [15]. The detailed measurements and analyses of oxygen contents were reported in our prior paper [15]. The deviations of the oxygen content (δ) of the $x = 0.04$ and 0.1 samples, decided by the oxygen/nitrogen determinator of LECO TC-300, are 0.016 and 0.01, respectively. The measurement of the average grain size was carried out using a JSM-6301F scanning electron microscope; the result showed that the average grain size was about 0.5 μm for $x = 0.04$ and 0.3 μm for $x = 0.1$. The magnetization measurements were carried out with a Quantum Design superconducting quantum interference device (SQUID) magnetometer in the temperature range of 5–300 K. Magnetoresistances were measured by the standard four-probe method. The micromagnetic properties were investigated using electron spin resonance (ESR) with a BRUKER-200D spectrometer.

3. Results and discussion

Figure 1(a) displays the curves of magnetization versus temperature at an applied field 1 T. The samples undergo a transition from a ferromagnetic state to a paramagnetic state with increasing temperature, from 5 to 300 K. Determining the transition temperature T_c as the peak value of the dM/dT versus temperature curve, the T_c obtained is about 201 and 240 K for $x = 0.04$ and 0.1.

In figure 1(b) the open triangles indicate the temperature-dependent resistivity $\rho(T)$ of $La_{0.96}Te_{0.04}MnO_3$ in zero magnetic field. One obvious feature is that the resistivity undergoes a transition characterized by the resistivity maximum from the metal to the insulator state with increasing temperature. The transition temperature T_{MI} is about 165 K. A similar transition occurs also in the sample of $La_{0.9}Te_{0.1}MnO_3$, marked by open circles in figure 1(b), but T_{MI} in zero field shifts to 210 K. Note that $T_{MI} < T_c$, which means that there exists an insulator phase in the long range ferromagnetic order state. On the other hand, the peak resistivity of the samples drops drastically when a high magnetic field, e.g. 4 T, is applied, as seen in figure 1(b) for $x = 0.04$, shown by a solid curve which indicates the existence of a colossal magnetoresistance effect.

Besides this, we have also performed a measurement of the magnetic field dependence of the resistivity $\rho(H)$ of $La_{0.96}Te_{0.04}MnO_3$ at various temperatures. The normalized curves of ρ–H are shown in figures 2(a) and (b). When the temperature is far below T_{MI}, e.g. 5 and 20 K, the resistivity of the sample drops sharply at first in the low field region, and then reduces linearly with increasing field in the high field range, as seen in figure 2(a), consistently with the results reported by Hwang *et al* [3] for polycrystalline ceramics $La_{2/3}Sr_{1/3}MnO_3$. Note

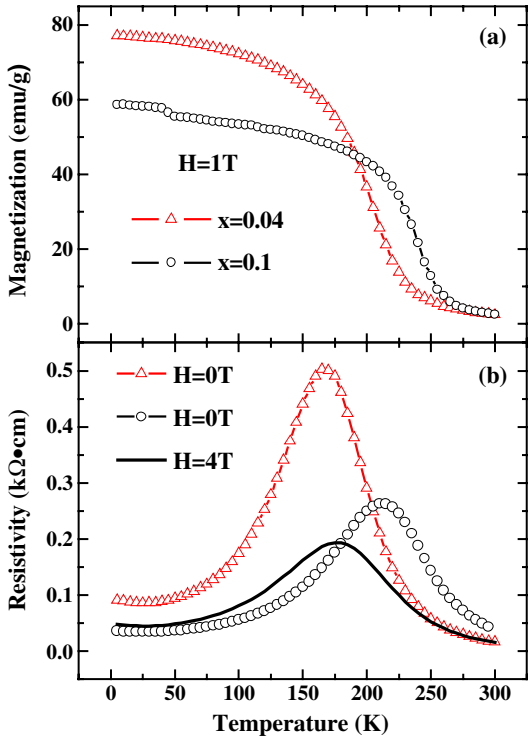

Figure 1. The temperature curves of magnetization and resistivity for $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$) in the temperature range of 5–300 K. (a) The magnetization at magnetic field 1 T. (b) The resistivity at zero field and 4 T marked by the solid curve.

that all the curves in figure 2(a) smoothly vary with increasing field, especially through the region of the magnetic domain rotation (see figure 5), which indicates that it is SPT rather than SDS that dominates the electron transport of samples in low field regime at $T \ll T_{MI}$ [3]. If we mark the field value where the line of the high field magnetoresistance intersects with the low field magnetoresistance as H_{LF}, the value obtained for H_{LF} is 0.27 T for 5 and 20 K, and the corresponding magnetoresistance ratio (MR) takes the values 33 and 31% for 5 and 20 K respectively; the definition is $MR = \Delta\rho/\rho_0 = (\rho_0 - \rho_H)/\rho_0$, where ρ_0 and ρ_H are the resistivity at zero and an applied field, as shown in figure 3 (the line is a guide to the eye). When the temperature is far above T_c, e.g. 250 K, a large LFMR with $MR = 18\%$ at 0.5 T is obtained. Even at room temperature (300 K), a weak but clear LFMR is still observed and the corresponding MR is about 3%. Undoubtedly, this result is very encouraging as regards future application of $La_{1-x}Te_xMnO_3$.

However, in the temperature range of T_{MI} to T_c, the MR is radically different from the case mentioned above, as shown in figure 2(b): no sharp drop in the curves of ρ–H at low fields appears; the resistivity at 170 and 200 K decreases smoothly with increasing field, even for the case at 135 K, below T_{MI}, which is similar to the results reported by Li *et al* for $Pr_{0.7}Pb_{0.3}MnO_3$ [16]. However, Li *et al* ignored the LFMR in the temperature range of far below T_c.

The variation of the resistivity $\rho(H)$ with the temperature mentioned above occurs also in the $x = 0.1$ sample (see figure 2(c), and the LFMR data are also shown in figure 3). When $T \ll T_{MI}$ (=210 K at zero field), e.g. 5 and 160 K, $La_{0.9}Te_{0.1}MnO_3$ exhibits a significant

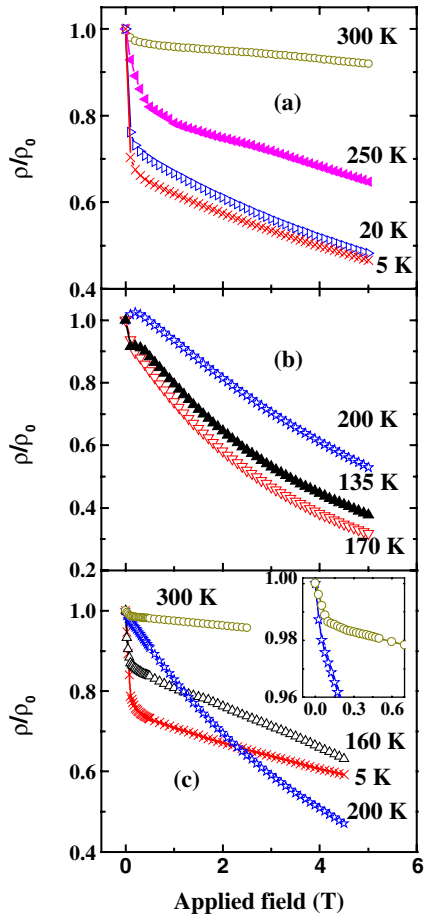

Figure 2. Magnetic field dependences of the resistivity normalized for $La_{0.96}Te_{0.04}MnO_3$ ((a) and (b)) and $La_{0.9}Te_{0.1}MnO_3$ (c) in the field range of 0–5 T at various temperatures. Panels (a) are for the cases $T \gg T_c$ and $T \ll T_{MI}$, panels (b) for the cases $T_{MI} < T < T_c$ and $T = 135$ K. The results in panels (c) at $T = 200$ and 300 K are locally zoomed in the inset.

LFMR effect. Moreover, the effect gradually weakens with increasing temperature and fades out until 200 K is reached. Compared with that for $La_{0.96}Te_{0.04}MnO_3$, the temperature regime where $La_{0.9}Te_{0.1}MnO_3$ displays a visible LFMR effect is wider in the case of $T < T_{MI}$. Similar to the case for $La_{0.96}Te_{0.04}MnO_3$, the LFMR effect of the $x = 0.1$ sample is observed again in the range of $T \gg T_c$. See the inset of figure 2(c); a large LFMR with MR = 2.3% at 0.1 T is obtained at room temperature.

Our observation gives a clear picture where the LFMR varies with the temperature. In order to further analyse the picture, we measured the ESR spectra versus temperature of the samples using a field-sweeping mode in the temperature range of 130–280 K for $x = 0.04$ and to 295 K for $x = 0.1$. The results are shown in figure 4. For $x = 0.04$, see figure 4(a), as the temperature is above 230 K, the ESR signals show a single symmetric peak with the Landé factor $g = 2.0$ nearly independent of the temperature, which is believed to be due primarily to paramagnetic Mn ions [17]. Below 230 K, the signal peak gradually broadens and shifts to low field with decreasing temperature. When $T < T_c$, a clear extra ferromagnetic resonance peak is observed. Up to $T < T_{MI}$, two signal peaks converge again, which means

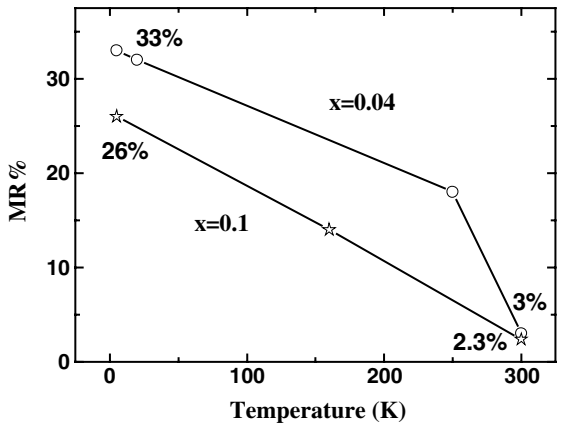

Figure 3. The low field magnetoresistance ratio at different temperatures for $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$). The solid line is a guide to the eye.

that the ferromagnetism of the sample is further enhanced. The ESR results above and previous analysis [15] indicate that, when the temperature is between T_{MI} and T_c, a paramagnetic phase and a ferromagnetic phase coexist in $La_{0.96}Te_{0.04}MnO_3$; this is called phase separation [18]. Obviously, the phase separation exists also in the sample with $x = 0.1$, as seen from figure 4(b).

According to ESR results, the magnetoresistance behaviours of the samples between T_{MI} and T_c are understandable due to the existence of the phase separation. On the other hand, the ESR results exhibit also the intrinsic magnetic inhomogeneity of the samples that was studied by Fäth [19] and Burgy *et al* [20]. As indicated in figure 4, with decreasing temperature, from room temperature to 130 K, the sample underwent a transition from a paramagnetic state to the coexistence of paramagnetic and ferromagnetic states, and then to a ferromagnetic state. Furthermore, the sizes of the different phases vary with temperature and field. Undoubtedly, the ferromagnetic phase of the samples will be enhanced when the temperature is below 130 K. However, as seen from the peak width and symmetry of the ESR signal at 130 K in figure 4, the ferromagnetic resonance signal possibly contains an antiferromagnetic signal, which is too weak and is submerged. The existence of the antiferromagnetic phase in the low compositional region of $R_{1-x}A_xMnO_3$ is also supported by Goodenough's work [21]. Additionally, we measured the magnetic field dependence of the magnetization $M(H)$ of $La_{0.96}Te_{0.04}MnO_3$ at various temperatures (shown in figure 5). Comparing to figure 4(a), we find that the sharp increase in magnetization at low temperature is associated with the sharp drop in resistivity. This result is consistent with that of Hwang *et al* [3], indicating an antiferromagnetic intergrain interaction. Thus, there may exist carrier-poor antiferromagnetic clusters in the sample that embed in the ferromagnetic matrix [22] at $T < 130$ K, leading to the large LFMR obtained at $T \ll T_{MI}$ due to the spin-polarized tunnelling between isolated ferromagnetic metallic clusters. This idea is also appropriate for the case of $T \gg T_c$, where the ferromagnetic clusters are surrounded by paramagnetic matrix, as indicated in figure 5. From figure 5, we see that the normalized magnetization varies nonlinearly with increasing field, and this is still the case even at $T = 230$ K. Besides this, when the samples approach the percolation threshold from the low temperature side, the large LFMR will gradually disappear, as shown at $T = 135$ K in figure 2(b). In contrast, away from the percolation threshold on the high temperature side, the LFMR will be obtained again, as seen for the case of $T = 250$ K; meanwhile, its value weakens with increasing temperature—see, e.g., the case for $T = 300$ K.

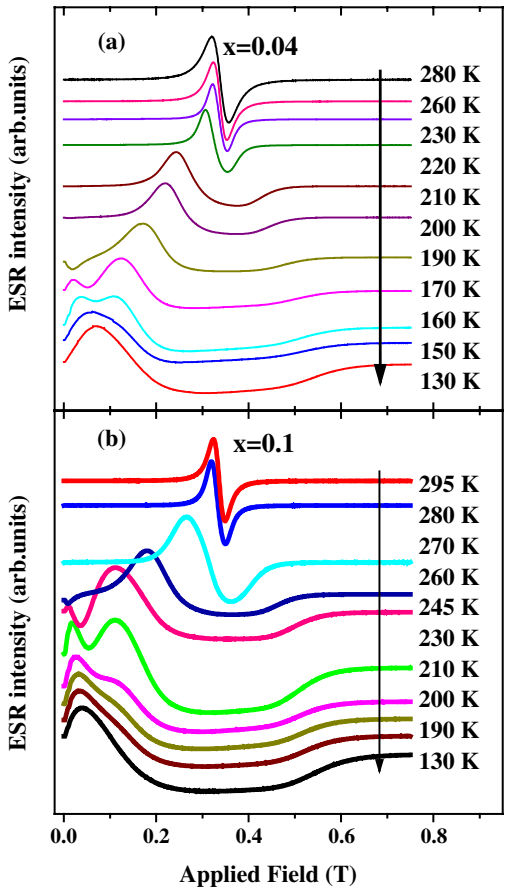

Figure 4. ESR spectra of $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$) measured in the field-sweeping mode at various temperatures. (a) $x = 0.04$; (b) $x = 0.1$.

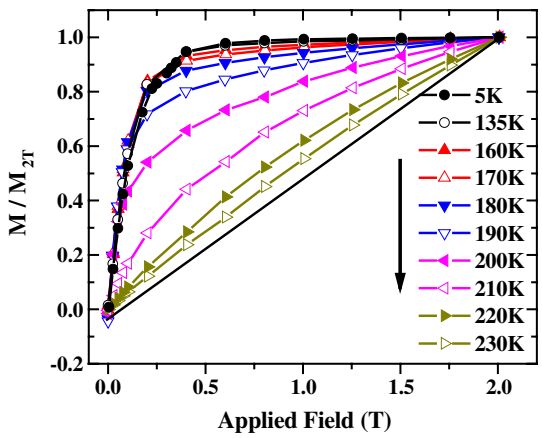

Figure 5. The magnetic field dependence of the magnetization normalized to the 2 T value for $La_{0.96}Te_{0.04}MnO_3$ in the temperature range of 5–230 K.

4. Conclusions

In summary, we have shown the temperature dependence of the LFMR for $La_{1-x}Te_xMnO_3$ ($x = 0.04, 0.1$) over the whole experimental temperature range of 5–300 K. A large LFMR effect was observed at $T \ll T_{MI}$ (MR $\sim$ 33% at $H_{LF} \sim 0.3$ T) and at $T \gg T_c$ (MR $\sim$ 18% at $H_{LF} \sim 0.5$ T), while in the temperature regime of $T_{MI} < T < T_c$, the resistivity of the samples changes smoothly with increasing field due to the existence of phase separation that probably masks the LFMR effect. The dominant contribution to the LFMR could stem from the spin-polarized tunnelling between isolated ferromagnetic clusters. Undoubtedly, our results will be helpful in further study of the LFMR effect of perovskite manganites.

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