CHIN.PHYS.LETT. Vol. 25, No. 10 (2008) 3773

Correlation between Electroresistance and Magnetoresistance in Slight Oxygen-Deficient $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ Polycrystalline Ceramics *

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(Received 25 April 2008)

A colossal electroresistance effect is found in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline ceramics with a very slight oxygen deficiency $\delta = 0.05$. The electroresistance behaviour of $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ ($\delta = 0.05$) displays the striking similarity to its counterpart magnetoresistance. It reveals that the boundary effect plays an important role in electroresistance and the same physical origin is likely responsible for both magnetoresistance and electroresistance effects in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystals.

PACS: 75.80.+q, 75.25.+z

Basically, divalent alkaline-earth doped manganite in perovskite-type materials is a strongly correlated electron system, and the physical properties are sensitive to external fields or the existing environmental conditions that even a small external stimulus can move its electronic state easily due to the delicate balance of energy resulting from the competition among the four main interactions, i.e. double exchange interactions, Hund coupling between Mn^{3+} e_g electrons and the core t_{2g} spins, electron-phonon interactions accompanied by the Jahn–Teller distortion, RKKY superexchange coupling and Coulomb repulsion.[1–3] The counterpart of magnetoresistance (MR) and electroresistance (ER) effect have increasingly received much attention since a huge ER was found in $Pr_{1-x}Ca_xMnO_3$ single crystal, in which ER was considered as a promising alternative for colossal magnetoresistance (CMR) in developing electronic devices with a fresh concept free from a conventional silicon technology.[4] In addition to the single crystal in which the charge-ordered (CO) insulating state was melt by electric-field to produce the colossal electroresistance (CER), CER was also found in thin films, particularly, in $La_{1-x}Ca_xMnO_3$ films with a coexistence of the CO insulating state and the ferromagnetic metallic state, due to the melting of the CO state equivalently by the high current density because of the very small transection area of thin film.[5–8] In this work, a remarkable ER was observed in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline ceramics with a slight oxygen deficiency. Such an ER effect is on very notable similarity to its counterpart MR, which can not be explained using the scenario of coexistence of the two competing phases or the melting effect on CO insulating state in single crystal or thin films. The striking similarity between ER and MR implies that the same physical origin is responsible for both the effects in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystals.

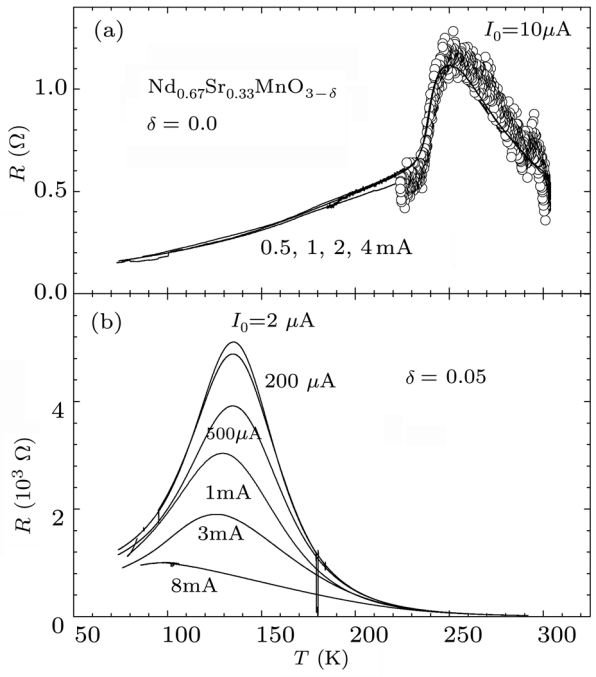


Fig. 1. $R(T)$ curves of $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline samples with constant current I_0 as a parameter for (a) $\delta = 0$ and (b) $\delta = 0.05$. A remarkable current-induced electroresistance effect occurs at $\delta = 0.05$ in a slight oxygen deficiency.

*Supported by the National Natural Science Foundation of China under Grant No 10774040, the Excellent Young Scientist Foundation of Hubei Province under Grant No 2006ABB032, and the Alexander von Humboldt Foundation.

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Ceramic $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ samples with $\delta = 0.05$ were prepared using the traditional ceramic techniques. The samples were placed in evacuated silica tubes together with some quantity of metallic tantalum, which served as an oxygen getter. The silica ampoules were fired at 900°C for 4 h and then cooled down to room temperature. For the reduced samples, the oxygen content has been determined from the mass loss of the sample. X-ray analysis has been performed using a DRON-3 diffractometer in the Cr–Ka-radiation mode. The samples show an orthorhombically distorted perovskite structure without any traces of foreign phases examined by x-ray diffraction. The electrical resistivity was measured by the standard four-point method.

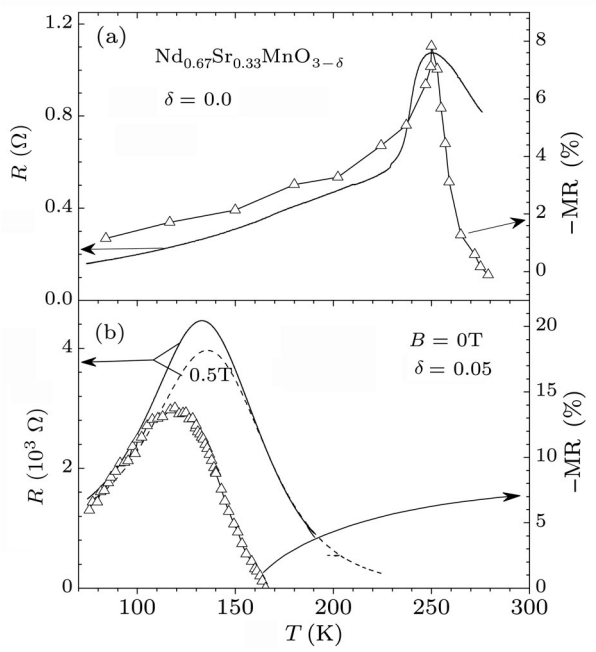


Fig. 2. $R(T)$ and MR(T) curves of $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline samples for (a) $\delta = 0$ under measuring current $I_0 = 1$ mA, magnetic field $B = 0.5$ T, and (b) $\delta = 0.05$ with $I_0 = 2\,\mu$A and $B = 0.5$ T. The magnetoresistance MR is defined as MR $= (R_H - R_0)/R_0$, R_0 the resistance at $B = 0$ and RH at $B = 0.5$ T, respectively.

Besides single crystal and thin films, CER was also observed in perovskites with grain boundaries, such as in $La_{2/3}Ca_{1/3}MnO_3$ bicrystal[9] and $La_{0.67}Ca_{0.33}Mn_{0.95}Mg_{0.05}O_3$ polycrystalline ceramics.[10] In our previous work, a remarkable ER was found in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystals with oxygen deficiency $\delta \geq 0.1$ below a characteristic temperature T_B marking a magnetic transition.[11] In this work, we report that even for a very slight oxygen-deficiency $\delta = 0.05$, $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ also shows a remarkable ER behaviour. Figures 1(a) and 1(b) show the current effect on electrical resistance for $\delta = 0$ and $\delta = 0.05$, respectively. For $\delta = 0$, i.e. the stoichiometric sample, the resistance is low and no ER occurs in the whole measuring range of temperature since all the $R(T)$ curves superpose together under the current loads. In this case, the grain boundary at $\delta = 0$ is conductive because of sufficient oxygen and electrical carriers can pass over the boundary as easily as the grain volume, so there is no ER taking place. However, with increasing oxygen deficiency to cause oxygen vacancies gathering inside boundary, the boundary becomes less conductive to induce the ER effect with enhancing the resistance due to the blocking potential of the barrier, as observed in Fig. 1(b). Such a boundary effect was also demonstrated by MR measurements, together with the temperature dependence of resistance in Fig. 2. MR curves of $\delta = 0$ (Fig. 2(a)) shows a very sharp peak at the same peak position of $R(T)$ around T_{MI} (metal-insulator transition temperature), similar to that in single crystal, while it shows a broad hump with the peak position shift to low temperature for $\delta = 0.05$ (Fig. 2(b)), which implies that the boundary plays an important role for the carrier transporting.[12]

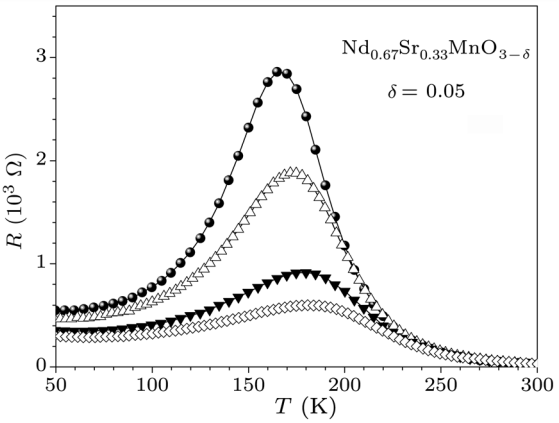


Fig. 3. $R(T)$ curves of $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline sample with constant field as a parameter for $\delta = 0.05$ measured by PPMS. The measuring constant current load is 0.5 mA.

More interesting, the CER curves is on striking similarity to its counterpart of CMR for $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystals with $\delta = 0.05$. Figure 3 shows the resistance dependence of temperatures under magnetic fields up to 5 Tesla. We can see that the two $R(T)$ curves in Figs. 1(b) and 3 are very similar in shape, both with the maximum of variation at around T_{MI} and decreasing quickly with temperatures apart from T_{MI} till $R(T)$ curves almost merging together resulting in a very small CER and CMR at both side of high and low temperatures. The similar variation of $R(T)$ with currents in Fig. 1(b) is also found for the sample under voltage loads. The striking similarity between the CMR and CER effects observed here suggests that both the effects may have the same physical origin, which we tentatively ascribe

to the magnetic-field induced by measuring current near the grain boundary of $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ ($\delta = 0.05$) polycrystals. For the conductance G of spin-dependent tunnelling across a barrier of boundary, it can be written as: $G = G_0(1+a\cos\theta_{12})$, where G_0 the conductance without boundary, $a \leq 1$ the spin polarization factor and θ_{12} the angle between the spin quantization axes at both sides.[13] Similar equations can be found for spin injection.[14] This approach may qualitatively explain the observed CER behaviour based on the creation of a magnetic field induced by measuring current. According to the equation, the boundary conductivity is dependent on the polarization direction of spins over the boundary. Under the consideration of the surprising similarity between CER and CMR in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$, it is reasonable to suppose that a magnetic field would be induced in the small region near the grain boundary as currents flow over. The spins around the boundary are expected to re-arrayed in accordance with the induced field, then θ_{12} is reduced dramatically at around T_{MI} so as to enhance the conductance G with respect to $G = G_0(1 + a\cos\theta_{12})$, therefore the ER effect is expected to produce consequently, just as observed in Fig. 1(b). The same correlation between CER and CMR was also reported in $La_{2/3}(Ca_{0.60}Ba_{0.40})_{1/3}MnO_3$ thin film.[15]

In summary, a remarkable ER has been found in $Nd_{0.67}Sr_{0.33}MnO_{3-\delta}$ polycrystalline ceramics and the striking similarity between CER and CMR implies that the same magnetic field origin is likely responsible for both the effects in the polycrystals.

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