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EFFECT OF Nd SUBSTITUTION ON ELECTRICAL TRANSPORT AND MAGNETORESISTIVE PROPERTIES OF $La_{2/3}Ba_{1/3}MnO_3$

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ABSTRACT

Polycrystalline semples of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ with $x = 0.0$, 1/6, 1/3, ½, 2/3, 5/6 and 1.0 have been prepared using solid state reaction. The metal-insulator transition (T_P) temperatures were determined by using the standard four-point probe resistivity measurement in a temperature range of 30K to 300K. T_p shifted to lower temperatures with the increase of Nd doping. On analyzing the data by using several theoretical models, it has been concluded that the metallic (ferromagnetic) part of the resistivity (ρ) below T_P fits well with the equation $\rho = \rho_0 + \rho_2 T^2 + \rho_{4.5} T^{4.5}$, indicating ρ_0 is due to the grain/domain boundary effects. A second term $\sim\rho_2 T^2$ appears might be attributed to electron–electron scattering and second-order electron–magnon scattering term $\sim\rho_{4.5} T^{4.5}$. The magnetoresistance (MR) effects are measured using the four point probe technique. The magnetoresistance defined as $MR\% = (R_o - R_H)/R_H \times 100$ was measured at magnetic fields $H \leq 1T$ at 90K, 150K, 250K and 300K. Overall, MR drops slowly when temperature rises. All doping concentration gives small variation range (~8.28% to ~56.53%). The highest MR value of ~56.53 % was measured at 1Tesla, at 100K for sample of $x = 1.0$.). At Low Field Magnetoresistance (LFMR), the highest gradient of MR is 125.35% MR/Tesla for sample x=0.0 at temperature 90K. The LFMR decreases prominently with increasing doping amount, while the HFMR is increased.

INTRODUCTION

The discovery of colossal magnetoresistance in mixed valent manganites has led to a resurgence of interest in this family of compounds. The general formula $Ln_{1-x}A_xMnO_3$ (Ln = rare earth and A = divalent alkaline earth cation) have created deal of interest because of their colossal magnetoresistance (CMR) behaviour. Many of the observed effects are due to the strong connection between magnetism and the insulator to metal transition special to these systems. Thus even at the insulating $LaMnO_3$ end, strong correlation, Hund's rule and Jahn teller effect are important. On substituting trivalent La with a divalent ion such as Ca or Ba, one d electron is transferred out from Mn^{3+} to oxygen, so that$La_{1-x}Ba_xMnO_3$ for instance has a fraction (1-x) of the Mn ions in the 3+ state and fraction x in the 4+ state. These systems have technological importance such as in sensor application, and especially for increasing data storage by increasing in sensitivity of hard disk drive read heads [1, 2]. A large low-field MR component has also been observed in polycrystalline manganite samples, where a disruption in the crystalline order at the grain boundaries induces a local spin disorder [3]. Using grain boundaries (GB's) to manipulate magnetic behaviour proves to be a simple method for enhancing the low-field sensitivity of these materials.

EXPERIMENTAL DETAILS

Polycrystalline of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ with x = 0, 0.167, 0.33, 0.5, 0.67, 0.833 and 1.0, were prepared via conventional solid-state reaction method. A well-mixed stoichiometric mixture of La_2O_3, Nd_2O_3, $BaCO_3$, $MnCO_3$ of 99.9% purities was mixed and grinded for 2 hours. The dried powder was heated at 900°C in air for 12 hours to produce a highly reactive powder. After calcinations, the black powdery mixture was reground, palletized, and sintered in air at 1300°C for 24 hours. DC four probe method with closed cycle helium refrigerator in the temperature range of 30 to 300 K was used to investigate the electrical properties.

RESULTS AND DISCUSSION

Figures 1(a)–(c) shows the temperature variation resistivity of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ samples. Samples with T_p show semiconducting transport behaviour above T_p and metallic behavior below T_p. The metal to insulator temperature, T_p shifted to lower temperature as Nd doping increases, which are >300, 248, 220, 198, 148, 112 and 70K for x = 0.0, 0.167, 0.333, 0.5, 0.667, 0.833 and 1.0, respectively.

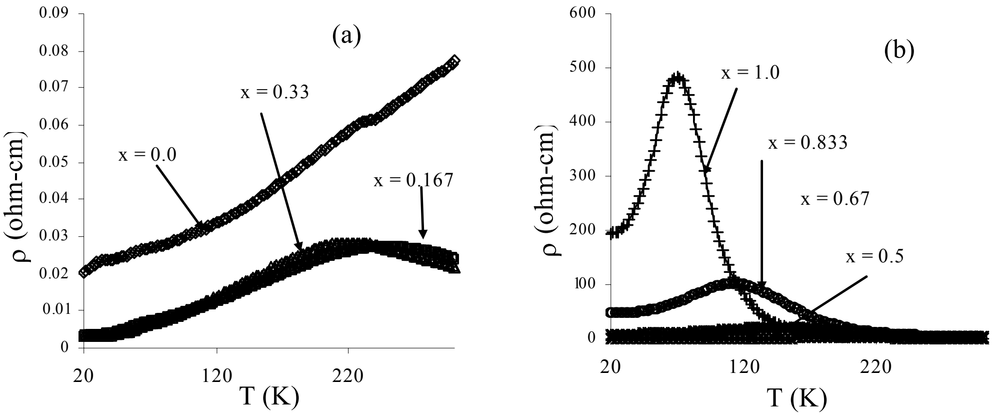

Figure 1(a)–(b): The temperature variation resistivity of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ samples;

Below T_p, the electronic conduction mechanism in the ferromagnetic metallic phase is generally understood according to double exchange theory. In this temperature regime, the resistivity data of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ samples fit quite well (see Figure 2) with the following expression:

$$\rho = \rho_0 + \rho_2 T^2 + \rho_{4.5} T^{4.5} \qquad (1)$$

where the first term ρ_0 corresponds to the resistivity arising due to domain, defect, grain boundary and domain walls. The second term $\sim\rho_2 T^2$ appears as a result of electron–electron and electron-phonon scattering mechanism (Ziese M, 2000), and the third term,

~$\rho_{4.5}T^{4.5}$with corresponds to the electron-magnon scattering (Rana D. S., *et. al*. 2004). Thus, the spin scattering cannot be neglected in the low temperature ($T < T_p$) regime as the measured data can be best explained by electron–magnon scattering.

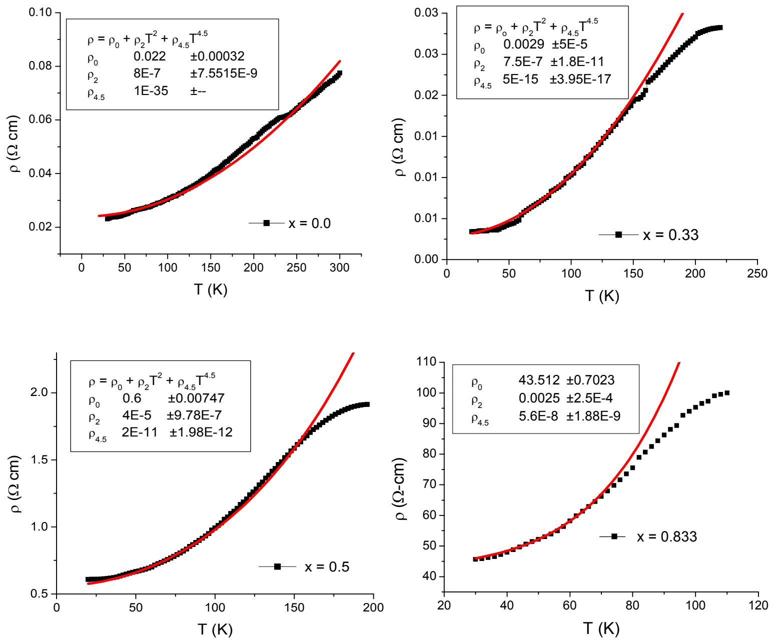

Figure 2: Replotted resistivity data for $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ system below T_p. Solid lines are the best fit to the equation $\rho = \rho_0 + \rho_2T^2 + \rho_{4.5}T^{4.5}$.

Table 1: Best fitted parameters obtained from the fitting of the low temperature resistivity data in the metallic regime with $\rho = \rho_0 + \rho_2T^2 + \rho_{4.5}T^{4.5}$

content, x	ρ_0 (Ω cm)	ρ_2 (Ω cm K^{-2})	$\rho_{4.5}$ (Ω cm $K^{-4.5}$)
0.000	0.0220	8.00E-07	1.00E-35
0.330	0.0029	7.50E-07	5.00E-15
0.500	0.5600	0.00004	2.00E-11
0.833	43.500	0.00250	5.60E-08
1.000	190.00	0.01000	2.70E-06

From Table 1, it is noted that the values of ρ_0, ρ_2 and $\rho_{4.5}$ increase with the increase of x. However, the decrease of temperature independent ρ_0 is more significant with x compared to that of ρ_2. As the doping increases, the size of the domain boundary decreases and ρ_0 becomes larger. The increase of ρ_2 and $\rho_{4.5}$ with x is due to spin fluctuation, consequently the bandwidth (b_p) become larger.

The MR defined as MR% = $(R_o - R_H)/R_H$ x 100% was measured at a magnetic field of H ≤ 1T at 90K, 150K, 250K and 300K as shown in Figure 3. All samples have increasing MR with increasing field with sample x=1.0 shows the highest MR around -

56.047% at 1T. The MR ratio at increasing rate with increasing Nd content. Overall, low concentration of doping ($0 \leq x \leq 0.5$) give two regions of MR effect significantly, which is low field MR (LFMR) exhibit a sharp drop at low field (0 to 0.1T) followed by a much slower linear decrease above 0.1T to 1T, so call high field MR (HLMR). However at high concentration (x=0.667- x=1.0), linear relationship is observed. At LFMR, the highest gradient of MR is 125.35% MR/Tesla for sample x=0.0 at temperature 90K. This value is very sensitive for low field application. For HFMR, the highest gradient of MR value is about 30.11% MR/Tesla at temperature 90K. The LFMR decreases prominently with increasing doping amount, while the HFMR increases. At room temperature, the highest gradient of LFMR is 63.6% MR/Tesla(x=0.0) decrease to 27.3% MR/Tesla(x=0.833). While, at HFMR, the gradient of MR is 5.26% MR/Tesla increase to reach the higher gradient of MR is about 11.70% MR/Tesla (x=0.5). The MR of a granular system consists of LFMR and HFMR, which are closely related to the surface effects of the grains. These effects increase gradually when decreasing grain size. The reduction of LFMR in Nd content samples closely relates to the improvement of the disordered state at the grain boundaries. Due to the lacking in electron tunneling probability at the grain boundaries, the spin dependent tunneling effect strengthens, and the LFMR decreases with increasing Nd content.

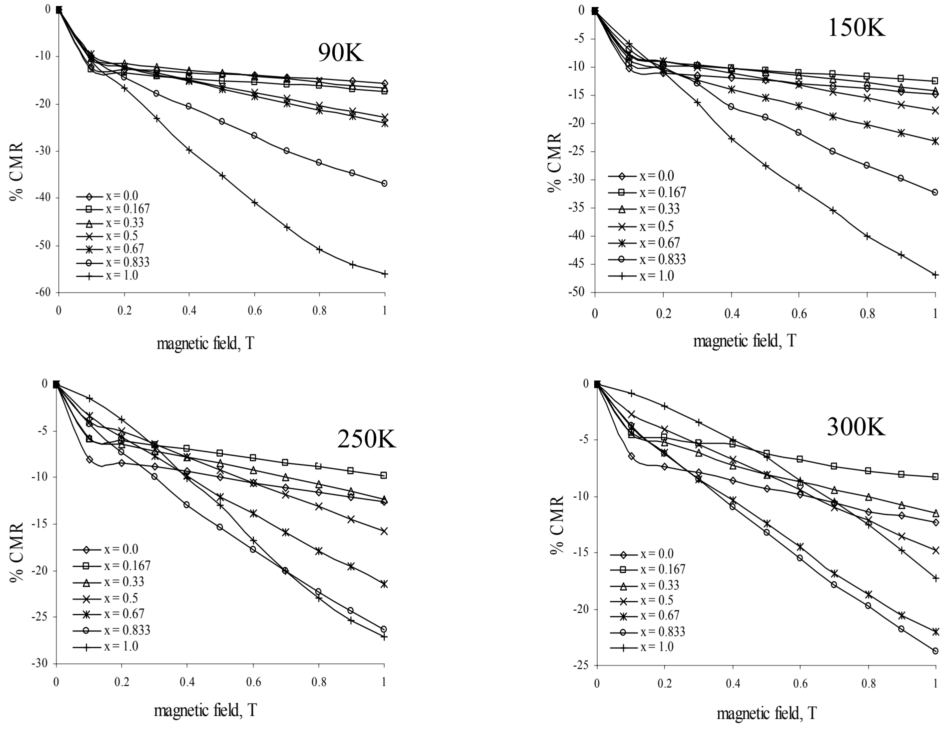

Figure 3: CMR curve of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ system as a function of applied magnetic field at 90K, 150K, 250K and 300K

Figure 4 exhibits MR of all samples increase when temperature are decrease. The higher MR value is about 56.54%, was obtained for sample of x=1.0. At this concentration, MR increased from 300K to 270K then, decrease slowly to 250K then increased to reach 90K. The manganese ion are ferromagnetically ordered below T_p; therefore, within a single magnetic domain, the e_g electrons transfer between Mn^{3+} and Mn^{4+} ions is easy. The pairs of Mn^{3+} and Mn^{4+} spins, which may not be parallel in the vicinity of domain wall boundaries, will act as a hindrance for electron transport. The magnetic domains tend to align along the field direction in the presence of sufficiently strong magnetic field. As a result, hopping of electrons become easy across the domain wall boundaries and the resistivity decreases, which in turn leads to significant MR at low temperature. Certain samples exhibit highest MR values close to their T_p values since this the property of mixed valence manganites. Close to T_p, the enhancements of MR are caused by the change of magnetic ordering from spin disorder of paramagnetic ordering to order spin of ferromagnetism ordering. Thus, the spin disorder scattering induced by a long range hopping correlation and its fluctuation, which is the possible origin of the CMR phenomena in doped manganites is reduced.

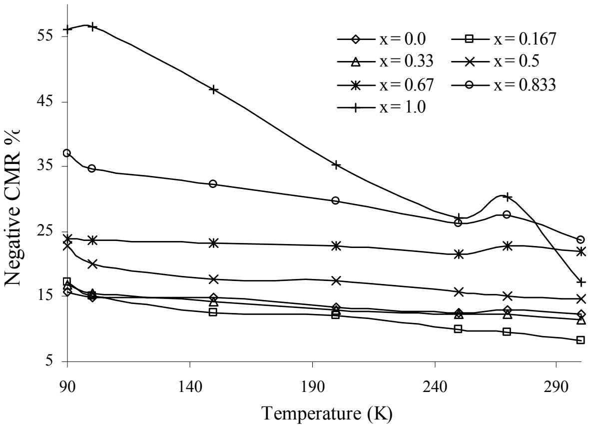

Figure 4: %MR of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ systems as function of temperature dependent

CONCLUSION

In conclusion, magnetotransport and magnetoresistive properties of $(La_{1-x}Nd_x)_{2/3}Ba_{1/3}MnO_3$ for $x = 0.0$, 1/6, 1/3, ½, 2/3, 5/6 and 1.0. Trivalent Nd doping in La-Ba-Mn-O systems drives the system towards lower conductivity and lower metal-insulator transition temperature region. Metallic conduction in these systems follows T^2 and $T^{4.5}$ dependence indicating the importance of electron–magnon scattering. MR drops slowly when temperature rises, except sample x=1.0. Sample x=1.0 exhibits a very high MR (~ -56.53%) at 1 Tesla measured at 100 K. Low concentration of doping ($0 \leq x \leq 0.5$) give two regions of MR effect, which is low field MR (LFMR) and high field MR (HLMR). At LFMR, the highest gradient of MR is 125.35% MR/Tesla for sample x=0.0 at temperature 90K. The LFMR decreases prominently with increasing doping amount, while the HFMR is increase.

ACKNOWLEDGEMENT

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