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HOPPING CONDUCTION IN $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$

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ABSTRACT

Electrical resistivity behaviour of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ compounds, prepared by the solid state reaction, have been investigated below the charge-ordering temperature to understand the mechanism of conduction. On analyzing the data by using several theoretical models, it is found that the metallic (ferromagnetic) part of the resistivity (ρ) (below T_P) fits well with the equation $\rho = \rho_0 + \rho_2 T^2$, where ρ_0 is due to the importance of grain/domain boundary effects, a second term $\sim\rho_2 T^2$ appears that might be attributed to the electron–electron scattering. In high temperature ($Tp<T<\theta_D/2$) paramagnetic insulating regime, the resistivity of samples obey the $T^{-1/4}$ law, characteristic of variable range hopping (VRH) model.

INTRODUCTION

La-*A*-Mn-O (A= a diavalent element; such as Ca, Ba, Sr, etc) is the one of widely studied materials of current interest showing semiconductor to metal transition [1]. However, the conduction mechanism in such mixed valence materials is a complex interplay between magnetic spin, charge ordering and also structure change. The Zener proposed that mechanism caused by a double exchange (DE) [2]. The basic process in DE is the hopping of d hole from Mn^{4+} to Mn^{3+} via oxygen. Therefore, polaronic hopping conduction mechanism is responsible for the conducting behviour in this system. Since lattice interaction is also involved, there is strong electron (hole) – phonon interaction is such compounds above the metal-insulator trasition (MIT). In addition, hopping mechanism of conduction corresponds to a very low mobility, since the electron jumps are associated with a weak overlap of wave-function tails from the neighboring. The variable range hopping theory has been widely used to explain the electric conduction process in manganites in the high-temperature regime is involved also equally semiconductor-like region. In this paper, we have studied the Al doping in place of Mn. The compounds $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ were prepared in single-phase form.

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EXPERIMENTAL DETAILS

Polycrystalline of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ (x = 0.0, 0.1, 0.2 and 0.3), were prepared via conventional solid-state reaction method. A well-mixed stoichiometric mixture of La_2O_3, Al_2O_3, $BaCO_3$ and $MnCO_3$ of 99.9% purities was mixed and grinded for 2 hours. The dried powder was heated at 900^{o}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^{o}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

Electrical resistivity of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ samples was measured in the temperature range 30 - 300K. Resistivity (ρ) versus temperature (T) plots for these samples are shown in Figure 1. All samples exhibit metal-insulator transition (MIT) with a maximum peak in the resistivity at a temperature defined as T_p. The values of T_p for each plot have been state in Table 2. T_p shifted to lower temperature as Al doping increases. This decrease, as expected, indicates that Al^{3+} does not take part in the double exchange mechanism. Therefore, the magnetic ordering takes place at lower temperature as x increases. The resistivity at T_p increases with Al concentration. For x = 0.3, the increment of resistivity is large. This is obvious because the replacing conducting regions of a conducting matrix by insulating regions.

To understand electronic conduction in $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ samples, at low temperature ($T < T_P$) the metallic behavior, we fitted the resisitivity data to Zener Double Exchange (ZDE) conduction law, given by;

$$\rho = \rho_0 + \rho_2 T^2 \qquad (1)$$

where the first term ρ_0 corresponds to the resistivity arising due to domain, grain boundary and other temperature independent scattering mechanism. The second term $\rho_2 T^2$ appears as a result of electron-electron scattering and electron–magnon scattering. 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.

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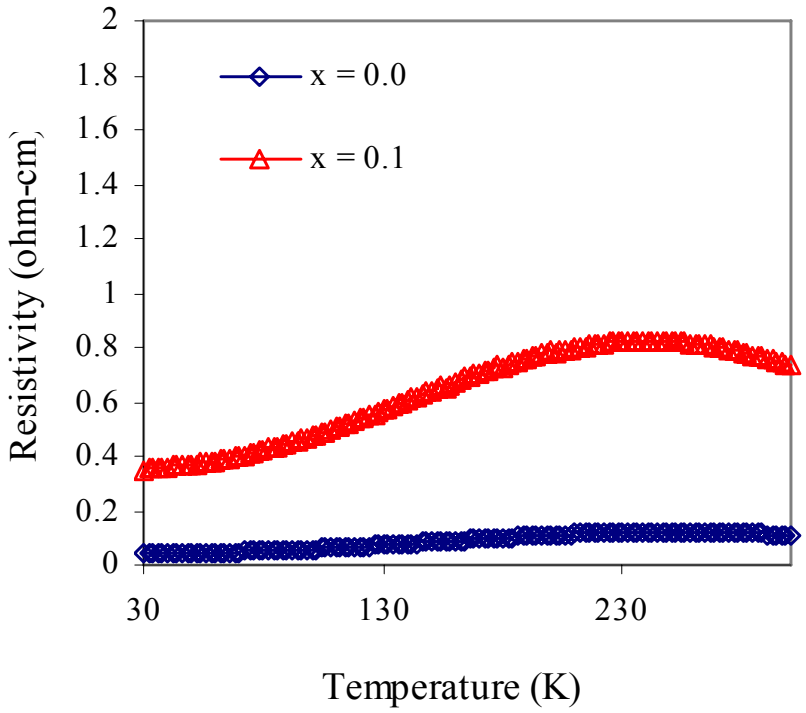

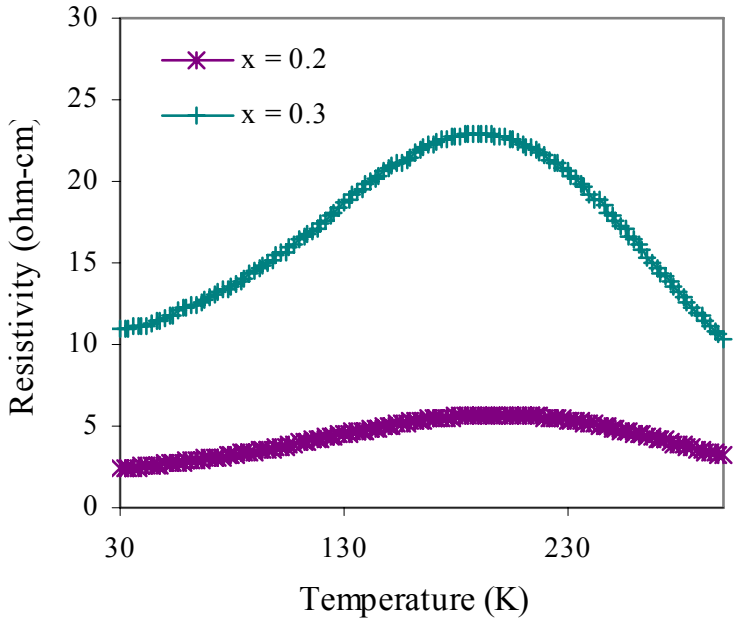

Figure 1 (a)(b): The temperature variation resistance of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$samples.

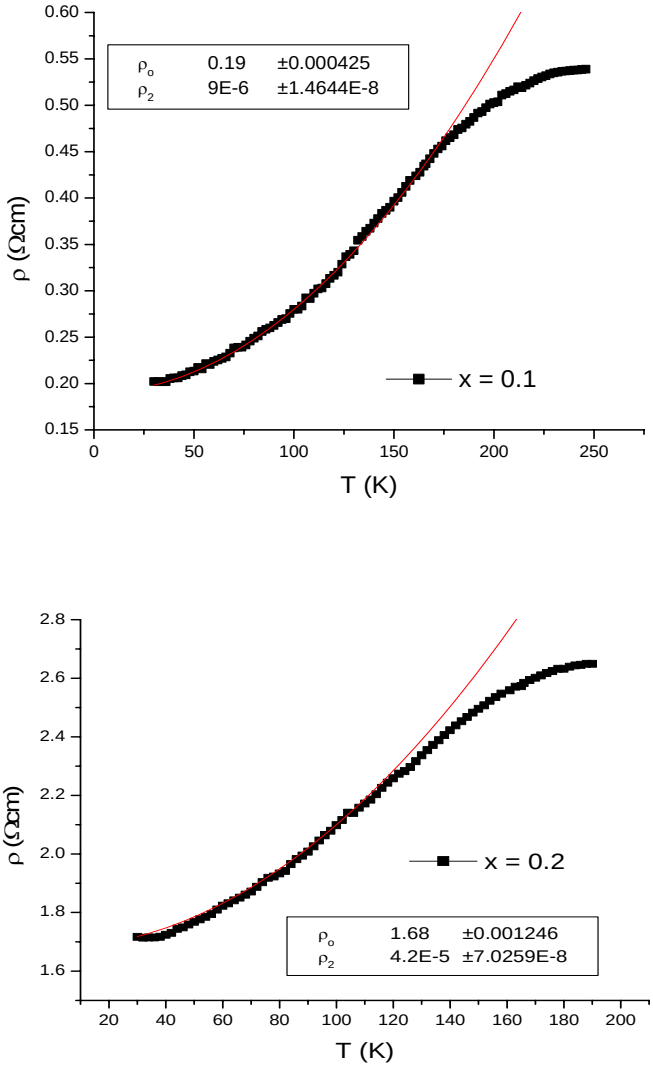

Figure 2: Replotted resistivity data showing T^2 dependence for $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ system below the respective T_p. Solid lines are the best fit to the equation $\rho = \rho_0 + \rho_2 T^2$.

The best fitted parameters are given in Table 1. It is noted that the values of both ρ_0 and ρ_2 increase with the increase of x. The increase of ρ_0 with x is suggested the increasing the grain boundary region. While, by the increase of ρ_2 with x is suggested due to the increment of the spin fluctuation. The spin fluctuation is caused by one-magnon scattering. Therefore, grain boundary plays a dominant role in the conduction process and it acts as the region of enhanced scattering center for conduction electron (A. Banerjee, 2002).

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Table 1: Best fitted parameters obtained from the fitting of the low temperature resistivity data in the metallic regime of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ manganites with $\rho = \rho_0 + \rho_2 T^2$.

concentration, x	ρ_0 (Ω-cm)	ρ_2 (Ω cm K^{-2})
0.00	0.04	1.50E-06
0.10	0.34	1.40E-05
0.20	2.32	1.50E-04
0.30	10.7	4.80E-04

It is well known that transport in semiconductor region ($Tp < T < \theta_D/2$) can be described by Variable Range Hopping (VRH) of charge carriers. Recently Jaime (1996) and Viret (1997) also applied VRH conduction mechanism in systems like Nd-(Sr,Pb)-Mn-O, La-Ca-Mn-O, etc [1]. The Mott mechanism of the variable range hopping (VRH) conductivity was obtained and has been utilized, given;

$$\sigma_{dc}=\sigma \exp(-T_0/T)^{1/4}, \qquad (2)$$

where, T_0 is a constant [$= 16\alpha^3/K_B N(E_F)$] and $N(E_F)$ is the density of states at the Fermi level which can be calculated from the slope of the plot log(σ) versus $T^{-1/4}$ curves as shown in Figure 3. Parameter α is the electron wave function constant, where considered as 2.22 nm^{-1} for the purpose of calculation. These also been used by Sayani *et al*. (2004) and Pal *et al*. (2001). $\theta_D/2$ values are also estimated, based on the graph $\theta_D/2 \approx T^{-1/4}$ where deviation from linearity occurs in the temperature region above T_p. The $\theta_D/2$ values are found to decrease systematically with increasing concentration, are given in Table 2.

Table 2: Values of T_p, T_0, $\theta_D/2$ and the density of states, $N(E_F)$ at Fermi level, from resistivity.

Content, x	T_p (K)	$\theta_D/2$(K)	T_0	$N(E_F)$($eV^{-1}cm^{-3}$)
0.00	252	262.237	6.176	5.015E+22
0.10	240	256.000	0.806	3.844E+22
0.20	198	218.024	0.258	3.989E+22
0.30	190	216.025	0.655	1.310E+22

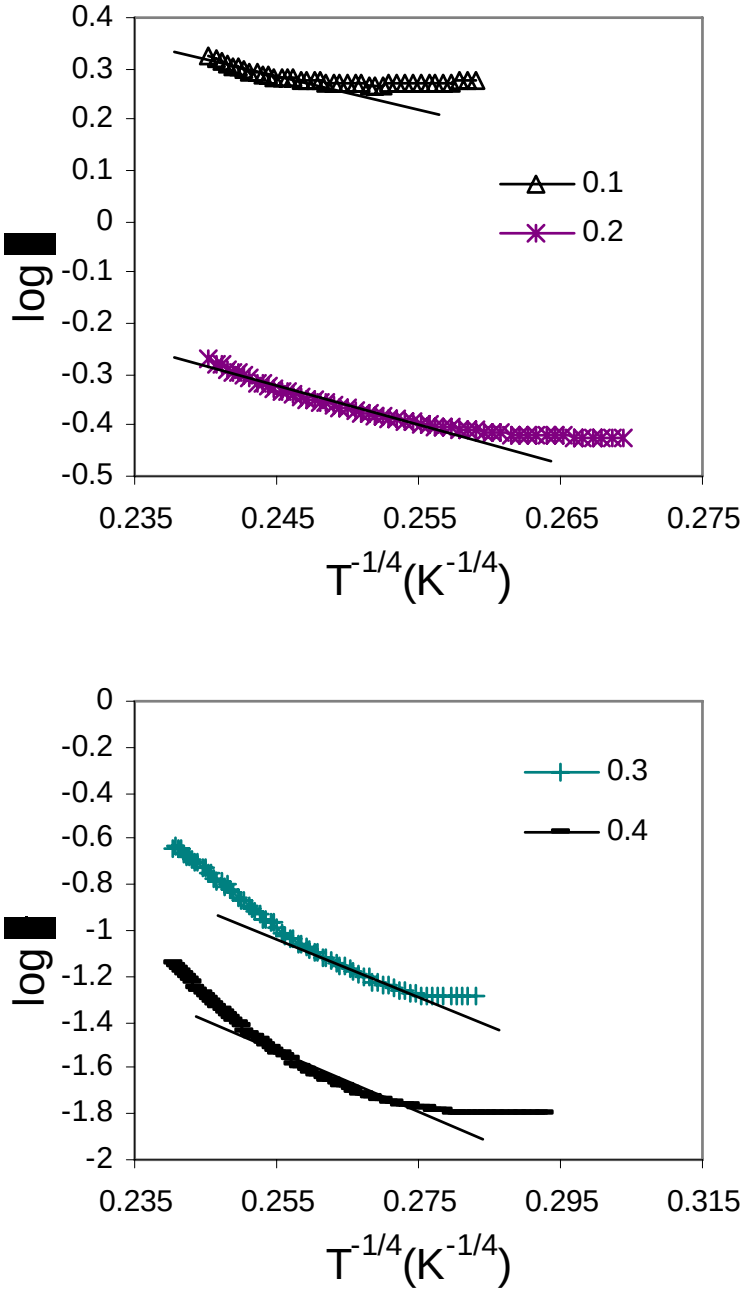

Figure 3: The fitting plot of σ (T) curves of $La_{1/2}Ba_{1/2}(Mn_{1-x}Al_x)O_3$ according to the VRH model. The dashed lines represent the experimental data.

Further T_0 values for each sample were calculated from the slopes of log (σ) vs. $T^{-1/4}$ plot. By using the T_0 values and the equation (3), $N(E_F)$, the density of states at the Fermi level for each material was also obtained. All the estimated parameters such as

$N(E_F)$, T_0 are found to be in agreement with, those reported in the literature for other manganites materials. T_0 values are found to increase enormously and continuously with increasing the concentration of Al. $N(E_F)$ are found to be decreasing continuously with the increasing the concentration. If the mobility of carrier reduces, this was suggested that the conduction of e_g band were decreased due to a strong electron-phonon coupling due to slowly fluctuating local Jahn-Teller distortion, permitting the formation of metallic state.

CONCLUSION

All samples show metal to insulator transition (MIT) at phase transition temperature (T_p). With the increase of Al content, T_p shifted to lower temperatures. The resistivity data (below T_P) fits well with the equation $\rho = \rho_0 + \rho_2 T^2$, where ρ_0 is due to the importance of grain/domain boundary effects, a second term $\sim\rho_2 T^2$ appears that might be attributed to the electron–electron scattering. The low temperature resistivity signifies the importance of electron-electron scattering and electron-magnon scattering; whereas the high temperature transport properties are mainly governed by the Variable Range Hopping (VRH) model. As the Al content is increased, the effective number density of electrons decreases due to the narrowing of the bandwidth. This will cause the reduction of conduction electron to hop to the neighboring sites.

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