Electrical conduction through bond percolation in $Nd_{0.67}Sr_{0.33}MnO_3$

M. Pattabiraman [a], G. Rangarajan [a,1]

[a] *Department of Physics, Indian Institute of Technology, Madras, Chennai 600 036*

Abstract

We report our analysis of the resistivity (ρ), data on $Nd_{0.67}Sr_{0.33}MnO_3$ polycrystalline samples as a function of preparative conditions using a bond percolation model for a random mixture of metallic and insulating regions, assuming polaronic transport above and below the metal-insulator (M-I) transition. Our analysis suggests that for oxygen defiecient compounds the M-I transition which occurs at a lower temperatre than the ferromagnetic transition arises due to a percolation of the metallic regions.

Key words: CMR, Manganites, percolation

1. Introduction

Recent NMR and X-ray Synchrotron measurements on the colossal magneoresistive manganite system of $Nd_{1-x}Sr_xMnO_3$ point to the existence of clusters with low carrier mobility in the metallic state well below T_C [1,2]. In addition, transport studies on the same system [3] and scanning tunnelling microscopy measurements [4]on $La_{1-x}SrxMnO_3$ ($\approx$0.3) point to the existence of metallic spin clusters above T_C. Thus the metal-insulator transition could be thought of as a gradual crossover process from one electronic state to the the other due to the competition between the double-exchange mechanism that tries to delocalize carriers and the electron-phonon coupling which tries to localize the carriers over an extended temperature range above and below T_C. The nature of such a crossover could be studied further without changing the chemical composition of the system by changing either the double-exchange strength-e.g. by hydrostatic pressure [5] - or the electron-phonon couping strength -say by creating oxygen vacancies resuling in broken Mn-O-Mn bonds [3]-. In this report we report our analysis of the temperature dependence of ($\rho(T)$) of $Nd_{0.67}Sr_{0.33}MnO_3$ for different processing conditions resulting in varying oxygen content in the lattice.This route is chosen in an attempt to account for the ρ(T) behaviour in these compounds with decreasing oxygen content. Polycrystalline powder of the compound was prepared by the standard solid state reaction route. For $\rho(T)$ studies (four probe method) five different samples were used: (1) 1500^{o}C sintered (code name: A1); (2) 1500^{o}C sintered, 10 hours Ar-annealed (A2); (3) 1500^{o}C sintered, 35 hours Ar-annealed (A3); (4) 1400^{o}C sintered, 5 hours O_2-annealed (A4) and (5) 1400^{o}C sintered, 5 hours Ar-annealed (A5). A laser ablated thin film of the same composition annealed first at 800^{o}C (N1A1) and 1000^{o}C (N1A2) was also used. The ρ(T) behaviour above and below T$_C$ of these samples was analyzed using a model which assumes that the electrical conduction occurs via a (bond) percolative process in a random network of resistances with two types of resistivities one more resistive than the other. At low temperatures, metallic behaviour dominates and at high temperatures the insulating behaviour dominates with a crossover at intermediate temperatures resulting in a metal-insulator transition. The expression for ρ(T) computed as a function of p -

[1] Corresponding author. Department of Physics, Indian Institute of Technology, Madras, Chennai 600 036. E-mail: rajan@physics.iitm.ernet.in

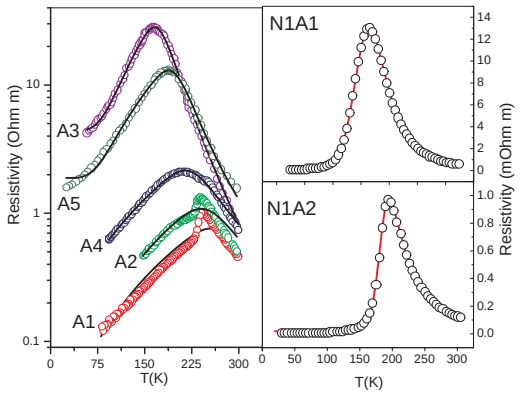

Fig. 1. Temperature dependence of resistivity of the bulk samples A1..A5 and thinfilms N1A1(p=0.39) and N1A2(p=0.42). The solid lines are the fits.

the metallic fraction using the bond percolation model is given by [6]

$$\rho = \frac{4\rho_1\rho_2}{((3p-1)\rho_1+(2-3p)\rho_2)+[((3p-1)\rho_1+(2-3p)\rho_2)^2+8\rho_1\rho_2]^{1/2}}$$

The simple case of a temperature independent p is considered and the temperature dependence of ρ_1 and ρ_2 are assumed to be independent of each other. The conduction via metallic regions is assumed to be due to small polarons and their corresponding resistivity is given by [7] $\rho_1 = \rho o' + E\omega_s/\sinh^2(\hbar\omega_s/2k_BT)$ and the conduction via non-metallic regions is assumed to be activated and is represented by the polaron hopping model [8] $\rho_2 = \rho_o T\exp(E_A/k_BT)$. ρ(T) plots of samples A1 through A5 with the fits obtained using the above equation are shown in fig. 1.

It is seen that the above equation can be used to fit ρ(T) over the entire temperature range for films, N1A1 and N1A2 and samples A3, A4 and A5. The fraction of metallic regions was found to increase with decrease in peak resistivity resistivity (table 1) and oxygen deficiency, pointing to a decrease in disorder. The fit equation does not reproduce the ρ (T) data of samples A2 and A1. When the sample becomes less resistive with a T_p closer to its T_C, the effect of magnetic transition appears in the ρ(T) curve as a sharp drop in resistivity at T_C (see ρ(T) plots for A1 and A2 in fig. 1) and the fit equation does not reproduce the ρ(T) behaviour around T_C. A temperature dependent p may have to be used to analyse the ρ(T) for samples with $T_p \approx T_C$. However a constant-p assumption seems to work for oxygen defiecient samples A3, A5 and A4 which have low T_p's (less than the observed T_C's - table 1) and higher resistivivity than samples A1 and A2. This suggests that the Mn-O-Mn bond disorder in these materials is large resulting in small poorly connected metallic clusters. A change in the size of the metallic clusters at T_C for these compounds does not bring about a prominent change in resistivity let alone the metal-insulator transition. Such a transition occurs only when a percolative path is established between the metallic clusters and the percoaltion model accounts for the large difference between T_C and T_p in oxygen deficient samples. The use of a percolative model with a constant 'p', is still useful as it provides a natural explanation for the decoupling of the metal-insulator and magnetic transitions in oxygen NSMO deficient samples. Thus the percolation via metallic regions determines T_p in oxygen deficient manganites and not the para-ferromagnetic transition.

1.0.0.1. *Table 1* The transition temperatures, resistivities at 300K and at the peak temperature, and the fraction (p) of metallic regions obtained from the fit.

Sample	T_C K	T_p K	$\rho(T_p)$ Ohmm	ρ(300K) Ohmm	p
A3 (1500 ^{o}C sintered) (35 hours Ar annealed)	215	165	28.5	0.8	0.4
A2 (1500 ^{o}C sintered) (10 hours Ar annealed)	235	238	1.3	0.5	0.62
A1 (1500 ^{o}C sintered) (as-prepared)	242	241	0.9	0.4	0.75
A5 (1400 ^{o}C sintered) (5 hours Ar annealed)	220	188	13	1.5	0.38
A4 (1400 ^{o}C sintered) (5 hours O_2 annealed)	229	213	2.1	0.7	0.59

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