Journal of Alloys and Compounds 616 (2014) 378–384

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

Review

Study of electrical transport and magnetoresistive properties of $La_{0.67-x}Dy_xPb_{0.33}MnO_3$ (x = 0.00, 0.10 and 0.15)

N. Zaidi [a,*], S. Mnefgui [a], A. Dhahri [a], J. Dhahri [a], E.K. Hlil [b]

[a] Laboratoire de la matière condensée et des nanosciences, Université de Monastir, 5019, Tunisia
[b] Institut Néel, CNRS et Université Joseph Fourier, B.P. 166, 38042 Grenoble, France

ARTICLE INFO

Article history:
Received 9 May 2014
Received in revised form 13 July 2014
Accepted 14 July 2014
Available online 1 August 2014

Keywords:
Manganites
Percolation theory
Magnetoresistance
Magnetic entropy change

ABSTRACT

We investigate the influence of dysprosium (Dy) doping on the magneto-electrical properties of LPMDy$_x$ compounds. The temperature dependence of electrical resistivity shows that all samples undergo a sharp metal–semiconductor (M–SC) transition at a temperature (T_{M-SC}), accompanying the ferromagnetic–paramagnetic transition. Metallic resistivity $\rho(T) = \rho_0 + \rho_1 T^2$ is observed below T_{M-SC}. Above T_{M-SC}, the electrical conductivity is dominated by adiabatic small polaron hopping model, inducing electrical resistivity in the paramagnetic region as $\rho_{PM}(T) = CT \exp\left(\frac{E_a}{k_B T}\right)$. A percolation theory is introduced to understand the properties of these mixed systems. We have also found that the estimated results are in good agreement with the experimental data. Above all, the magnetoresistance (MR) study showed a peak which has a great value around the metal–semiconductor (M–SC) transition temperature. The dependence of resistivity on the temperature and magnetic field data is used to deduce the magnetic entropy change.

Contents

1. Introduction

The perovskite manganites of a general formula $Re_{1-x}A_xMnO_3$ (Re: trivalent rare earth element and A = alkaline earth) have become an interesting object due to their important electrical and magnetic properties such as the colossal magnetoresistance (CMR) [1–3] and the magnetocaloric effect (MCE) [4–6]. The most important feature of these materials is the strong correlation between structure, transport and magnetic properties as well as their potential in technological applications [7–9]. The strong correlation between magnetic and transport properties in these materials have been explained within the framework of the double-exchange (DE) model. The fundamental process in the double exchange is the hopping of d hole from $Mn^{4+}(t_{2g}^3 eg^0)$ to $Mn^{3+}(t_{2g}^3 eg^1)$ via oxygen (Mn^{4+}–O^{2-}–Mn^{3+}), so that Mn^{4+} and Mn^{3+} ion exchange takes place. However, recent studies have shown that the double exchange (DE) interaction, between Mn^{3+} and Mn^{4+} ions, cannot explain alone the behaviors observed in these systems and it suggested that other effects like the average A-site cationic radius ‹r_A›, the grain boundaries [10,11] and the oxygen deficiency [12,13] can play a crucial role for better explanation. Experimentally, manganese oxides exhibit a metal–semiconductor (M–SC) transition accompanied by a ferromagnetic–paramagnetic

* Corresponding author. Tel.: +216 97 543 131
E-mail address: zaidinadia77@yahoo.Com (N. Zaidi).

http://dx.doi.org/10.1016/j.jallcom.2014.07.113
0925-8388/

(FM–PM) transition near the Curie temperature T_C. The metallic behavior is usually described in terms of electron- phonon coupling [14,15] in relation with the formation of polarons in the paramagnetic state, in addition to the double exchange coupling (DEC), recently used to explain the experimental results including the transport mechanisms [16,17]. Some models can explain the electronic transport mechanism in manganite materials. However, most of them are only applied to fit the prominent change of the ρ–T curves in a finite temperature region (above or below T_C). A phenomenological percolation model, based on phase segregation which is proposed by Li et al. [18,19], has been successfully used to explain the transport mechanism in the whole temperature range. Following this mechanism, the electrical resistivity at any temperature is determined by the change of the volume fractions of both regions (paramagnetic and ferromagnetic). Further, the double exchange theory [20,21] qualitatively shows a correlation between ferromagnetism and electrical conductivity in some manganites [22,23]. Xiong et al. also found a strong correlation between resistivity and magnetic-entropy change (ΔS_M) [24]. So, we can calculate ΔS_M based on the electrical measurement.

According to the literature, the effect of substitution at A-site of perovskite manganites produces a strong change in the physical properties, essentially the Curie temperature. It was reported that the partial substitution of Pr^{3+} by Bi^{3+} in $Pr_{0.6-x}Bi_xSr_{0.4}MnO_3$ causes sharp changes in the electrical properties. Furthermore, this substitution by Bi^{3+} decreases the Curie temperature and enhances the resistivity of the samples [25]. Dinesh et al. [26] showed also that the doped $La_{0.85-x}Sm_xK_{0.15}MnO_3$ compounds with $0.05 \leqslant x \leqslant 0.15$ exhibit lower Curie temperature (T_C) and metal-isolant transition temperature ($T_{M\text{-}I}$). The electrical below $T_{M\text{-}I}$ of all polycrystalline $La_{0.85-x}Sm_xK_{0.15}MnO_3$ attributes to electron–electron, electron–phonon and electron-spin fluctuation scattering processes, while in the high temperature insulating regime, the conduction is due to adiabatic small polaron. In the present work, in order to decrease the critical temperature of the parent compound $La_{0.67}Pb_{0.33}MnO_3$ (T_C = 360 K) [27], we substituted Lanthanum (La) by dysprosium (Dy). From the relationship between the theoretical and the experimental data, we will make clear the role of the trivalent magnetic ion (Dy) in the ferromagnetic–paramagnetic and metallic–semiconductor transitions. Analyses of resistivity based on the percolation theory as well as the investigation of the magnetoresistance effect in our samples are reported. Finally, we have deduced the magnetic entropy change ($-\Delta S_M$) from resistivity vs. temperature data measured at various magnetic fields.

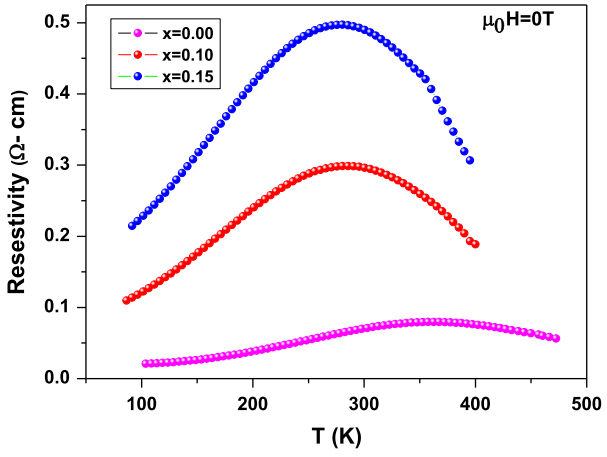

Fig. 1. Resistivity vs. temperature plot of $La_{0.67-x}Dy_xPb_{0.33}MnO_3$ (x = 0.00, 0.10 and 0.15) at zero magnetic field.

2. Experimental

Dy-doped lanthanum manganites with a chemical composition ($LPMDy_x$) (x = 0.00, 0.10 and 0.15) were prepared using the standard ceramic reaction at high temperatures. The mixtures of the respective oxides were calcinated at a temperature of 1373 K and then were pressed and sintered in air at 1673 K for 24 h. The structural and magnetic properties were investigated in a previous work [28]. Electrical resistivity as a function of temperature was measured by a conventional four-probe method in the temperature interval from 50 to 400 K. In order to analyze the resistivity data of high temperature region, $T > T_{M\text{–}SC}$, we have used the adiabatic small polaron-hopping model (SPH). It is worth mentioning that the most rapid motion of a small polaron occurs when the carrier hops each time the configuration of vibrating atoms in an adjacent site coincides with that in the occupied site. Henceforth, the charge carrier motion within the adiabatic regime is faster than the lattice vibrations and the resistivity for SPH follows: $\rho_{PM}(T) = CT \exp\left(\frac{E_a}{k_B T}\right)$. The $T < T_{M\text{–}SC}$ experimental data were fitted by $\rho(T) = \rho_0 + \rho_2 T^2$. The percolation model, based on phase segregation has been used successfully in whole measured temperature where resistivity can be written in the following way: $\rho(T) = \rho_{FM} f + \rho_{PM}(1 - f)$.

3. Results and discussion

3.1. Electrical behavior

The temperature dependence of the resistivity ($\rho(T)$) of $LPMDy_x$ (x = 0.00, 0.10 and 0.15) polycrystalline samples was measured in the 50–400 K temperature range in zero magnetic field. This is shown in Fig. 1. Taking the sign of the temperature derivative of the resistivity ($d\rho/dT$) as a criterion, we found that samples exhibit a metallic behavior ($d\rho/dT > 0$) at low temperature ($T < T_{M\text{–}SC}$) and become semiconductor-like ($d\rho/dT < 0$) above the temperature $T_{M\text{–}SC}$, where $T_{M\text{–}SC}$ is the temperature of the maximum value of resistivity ρ_{max}. For samples x = 0.00, 0.10 and 0.15, the transition temperature $T_{M\text{–}SC}$ occurs at 335 K, 295 K and 275 K respectively (Table 1), which are close to their Curie temperatures T_C (T_C = 360 K, 317 K and 291 K), indicating strong correlations between the magnetic and electrical properties in the $LPMDy_x$ samples. We can therefore, define the ferromagnetic-metallic-like and the paramagnetic-semiconductor like behaviors as a function of temperature. The electrical resistivity increases with Dy concentration. This could be associated to a successive substitution of Dy^{3+} at A-site (La^{3+}), which reduces the value of $\langle r_A \rangle$. Consequently, the ferromagnetic double exchange (FMDE) mechanism is weakened due to a slight distortion of the MnO_6 octahedra [29]. It causes the deviation of Mn^{3+}–O–Mn^{4+} bond angle from 180° and affects the e_g electron hopping rate between Mn^{3+} and Mn^{4+}. As a result, the resistivity increases and then the transition becomes sharper [30]. This can be explained by a co-existence of metallic and semi-conductor behavior. That is due to the fact that the electrical resistivity of our materials depends on the respective volume fraction of $La_{0.67-x}Dy_xPb_{0.33}MnO_3$ (x = 0.00, 0.10 and 0.15) phases.

Fig. 2 shows the effect of a 2 and 5 Tesla magnetic field on ρ ($T_{M\text{–}SC}$) curves of x = 0.00, 0.10 and 0.15. The obtained values of $T_{M\text{–}SC}$ and $\rho(T_{M\text{–}SC})$ are summarized in Table 1. The decrease of resistivity when the magnetic field increases may be due to the fact that the applied magnetic field induces the delocalization of charge carriers. As a result, the ferromagnetic metallic state may suppress the paramagnetic-semiconductor regime. In fact, randomly oriented moments of particles can be aligned by an external field. This

Table 1
Metal–semiconductor transition temperature (T_{MSC}) and maximum of resistivity at T_{MSC}.

	T_{MSC} (K)			P (Ω-cm) at T_{MSC}		
	H = 0 T	H = 2 T	H = 5 T	H = 0 T	H = 2 T	H = 5 T
x = 0.00	335	332	330	0.101	0.079	0.050
x = 0.10	295	292	290	0.299	0.258	0.194
x = 0.15	275	273	268	0.499	0.445	0.421