# Magnetic and transport properties of $La_{0.7-x}Pr_xPb_{0.3}MnO_3$

Y. C. Chen [a,1], S. L. Young [a], H. Z. Chen [b], Lance Horng [c], J. B. Shi [d]

[a] *Department of Electrical Engineering, National Sun Yat-Sen University, Kaohsiung 804, Taiwan.*
[b] *Department of Electrical Engineering, Hsiuping Institute of Technology, Taichung 412, Taiwan.*
[c] *Department of Physics, National Changhua University of Education, Changhua 500, Taiwan.*
[d] *Department of Electronic Engineering, Feng-Chia University, Taichung 407, Taiwan.*

**Abstract**

The substitution of Pr on the La site in $La_{1-x}Pr_xPb_{0.3}MnO_3$ has been studied in order to probe into the physical mechanisms of colossal magnetoresistance (CMR) behavior. The magnetic order changes from a long-range order with $x$=0.0 to a short-range nature with $x$=0.7. The ferro-paramagnetic (FM-PM) and metal-insulator (M-I) transition temperature values decrease as Pr content increase. The highest MR ratio values is 16.9% for $x$=0.0 against 41.6% for $x$=0.7 in an applied field 1 tesla. It is suggested that the mean ionic radius of the La-site ions may play an important role on the CMR effect.

*Key words:* colossal magnetoresistance; ferromagnetic-paramagnetic; metal-insulator

Perovskite manganese oxides, $Ln_{1-x}A_xMnO_3$ (Ln=La, Nd, Pr, Sm, etc., and A=Ca, Sr, Ba, Pb, etc.), have recently attracted considerable attention because of the rich variety of crystallographic, magnetic, and electronic phases [1–3]. Recent researches of the phenomenon of CMR in rare earth manganites have clearly suggested that these oxides exhibit a metal-insulator (M-I) transition at the temperature $T_P$ which is close to the ferromagnetic transition temperature $T_C$. There are two important structural parameters, Mn—O bond length and Mn—O—Mn bond angle, in the CMR compounds. The motion of the $e_g$ electron from $Mn^{3+}$ to $Mn^{4+}$ can be strongly influenced by the mean ionic radius of the La-site ions which exhibits a close relationship between the bond length and bond angle of $Mn^{3+}$-$O^{2-}$-$Mn^{4+}$ and the electron band width [4,5]. Therefore, the substitution of smaller lanthanides for La gives rise to a decrease of tolerance factor $t$, defined as $t=(r_A+r_O)/\sqrt{2}(r_B+r_O)$ of $ABO_3$, a bending of the Mn–O–Mn bond from 180$^o$ to (180$^o$-$\varphi$) [6], causes the narrowing of the $e_g$ electron bandwidth [7,8], suppress the DE interaction and decrease the $T_C$. In this work, the authors intend to illustrate the substitution effect of La by smaller ion Pr in the $La_{0.7-x}Pr_xPb_{0.3}MnO_3$.

Polycrystalline $La_{0.7-x}Pr_xPb_{0.3}MnO_3$ specimens were synthesized by conventional solid-state reaction method. The structure and phase purity of the samples were examined by powder $x$-ray diffraction using Cu-K$\alpha$ radiation at room temperature. All the specimens were single phase with no detectable secondary phases. The magnetization and resistivity measurements were performed by a Quantum Design MPMS SQUID magnetometer.

In order to get insight into the spin order and magnetic behavior, the ZFC-FC magnetization curves were measured at a field of 100 Oe as shown in figure 1. This is one of method generally used to characterize the spin ordering behavior. For sample $x$=0.0, the almost overlap of the ZFC-FC magnetization curve suggests a ferromagnetic long-range spin ordering. Conversely, the irreversibility between the ZFC and FC magnetization curves for other samples is clearly seen below $T_C$. The ZFC-FC curves display the irreversibility and $\lambda$-shape

[1] Corresponding author. Present address: Department of Electrical Engineering, National Sun Yat-sen University, 70 Lienhai Rd., Kaohsiung 804, Taiwan. E-mail: ycc@ee.nsysu.edu.tw

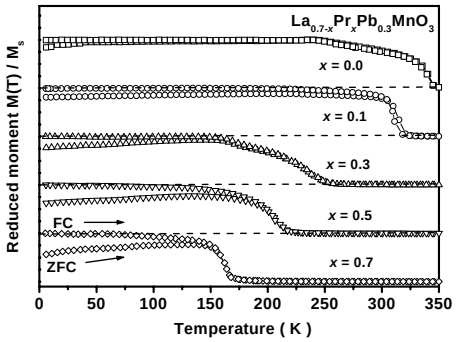


Fig. 1. The temperature dependence magnetization measurements at a low field of 100 Oe.

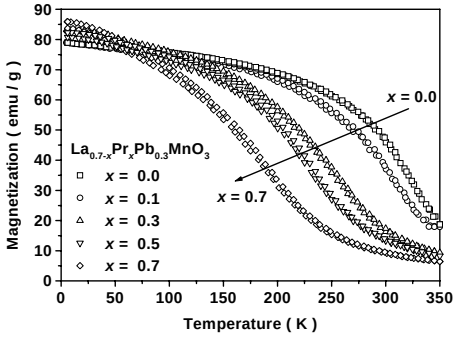


Fig. 2. The temperature dependence of magnetization at 5 T applied field for all compositions. Arrow is a guide for the eyes.

traces, indicating the existence of a short-range spin ordering. Therefore, the $La_{0.7}Pb_{0.3}MnO_3$ has long-range spin ordering, while $Pr_{0.7}Pb_{0.3}MnO_3$ has short-range one. The reason for the variation is the introduction of small Pr ions to the La-site that leads to a large deformation of the octahedrons, bend the Mn—O—Mn bonds and, consequently, weaken the DE interaction between $Mn^{3+}$ and $Mn^{4+}$ in these perovskites.

Figure 2 shows the temperature dependence of magnetization at 5 T for all compositions. The transition temperatures ($T_C$), defined as the temperature where the value of $dM(T)/dT$ reaches the maximum value, decrease as Pr content increase. This fact is in good agreement with the introduction of small ionic radius of the Pr ion, the corresponding distortion of the perovskite structures induced by smaller $t$, the abatement of DE spin coupling and, consequently, the decrease of $T_C$. The saturated magnetizations ($M_S$), obtained from the saturated value of the magnetizing curve at 5 K and 5 T, increase with Pr content. This can be illustrated in term of canted spins between Mn and magnetic Pr. The magnetic ions $Pr^{3+}$ with $f$-shell electrons are gradually aligned with the manganese as the field increases at low temperature. Thus, the rare earth magnetic ions contribute to an additional value to the total moment for Pr-rich compositions.

Table 1
Values of tolerance factor $t$, ferromagnetic transition temperature $T_C$, saturation magnetization moment $M_S$, M-I transition temperature $T_P$ and MR ratio.

| Composition | $t$ | $T_C$ (K) | $M_S$ ($\mu_B$) | $T_P$ (K) | MR (%) |
|---|---|---|---|---|---|
| $x$=0.0 | 0.9915 | 332 | 78.90 | 329 | 17.09 |
| $x$=0.1 | 0.9893 | 316 | 79.08 | 312 | 17.74 |
| $x$=0.3 | 0.9850 | 234 | 82.36 | 224 | 23.35 |
| $x$=0.5 | 0.9807 | 202 | 83.18 | 151 | 36.14 |
| $x$=0.7 | 0.9764 | 163 | 85.94 | 94 | 41.27 |

The temperature $T_P$ where metal-insulator transition takes place and MR% defined as MR%=[$\rho$(H=1T)-$\rho$(H=0T)]/$\rho$(H=1T) for all samples were listed in table 1. $T_P$ is reduced when Pr doping content is increased from 331K for $x$=0.0 to 94K for $x$=0.7. Resistivity and magnetoresistance ratio increase manifestly as Pr content increases. This is an expected result that synthesizes the relation between the lattice effect induced by substitution and CMR effect. By gradual substitution of Pr for La, the deformation of crystal structure increases and the bond angle of Mn—O—Mn decreases. The reduction of bond angle is accompanied with a weaker $e_g$—$2p_\sigma$—$e_g$ overlaping, a narrower $e_g$ bandwidth and smaller carrier mobility. In addition, the distortion leads to the increase of the canting angle of moments and resistivity. Thus, resistivity increases as Pr doping increases. Due to the parallelity of moments with an external field, the resistivity will be reduced obviously. Therefore, MR% increases from 17.1% for $x$=0.0 to 41.3% for $x$=0.7 around $T_P$ as Pr doping increases.

The magnetic and transport properties of Pr doped $La_{0.7}Pb_{0.3}MnO_3$ have shown in this work. The physical properties vary systematically by changing the Pr doping content $x$. While increasing $x$, the $T_C$ and $T_P$ decrease, and maximum MR% around $T_P$ increases as well. In conclusion, these effects are attributed to the crystal deformation induced by the substitution of Pr onto La-site.

## References

[1] K. Ghosh *et al.*, Phys. Rev. B **59** (1999) 533.

[2] J. B. Goodenough, J. Appl. Phys. **81** (1997) 5330.

[3] J. P. Zhou *et al.*, Appl. Phys. Lett. **75** (1999) 1146.

[4] J. Fontcuberta *et al.*, J. Appl. Phys. **81** (1997) 5481.

[5] S. L. Young *et al.*, J. Mag. Mag. Matter **239** (2002) 70.

[6] R. Mahendiran *et al.*, Phys. Rev. Lett. **75** (1995) 914.

[7] Y. Tomioka *et al.*, Appl. Phys. Lett. **70** (1997) 3609.

[8] I. O. Troyanchuk *et al.*, Phys. Rev. B **58** (1998) 2422.