

# Evolution of magnetic properties and canted spin behavior of the $\text{La}_{0.7-x}\text{Sm}_x\text{Pb}_{0.3}\text{MnO}_3$ manganites

H. Z. Chen <sup>a,1</sup>, S. L. Young <sup>b</sup>, Y. C. Chen <sup>b</sup>, Lance Horng <sup>c</sup>, J. B. Shi <sup>d</sup>

<sup>a</sup>Department of Electrical Engineering, Hsiuping Institute of Technology, Taichung 412, Taiwan.

<sup>b</sup>Department of Electrical Engineering, National Sun Yat-Sen University, Kaohsiung 804, Taiwan.

<sup>c</sup>Department of Physics, National Changhua University of Education, Changhua 500, Taiwan.

<sup>d</sup>Department of Electronic Engineering, Feng-Chia University, Taichung 407, Taiwan.

---

## Abstract

The magnetic and transport properties of mixed-valence manganites  $\text{La}_{0.7-x}\text{Sm}_x\text{Pb}_{0.3}\text{MnO}_3$  ( $x=0, 0.1$  and  $0.3$ ) are investigated. The  $x=0$  phase is indexed in a rhombohedral structure ( $R\bar{3}c$ ) while the rest exhibit orthorhombic symmetry ( $Pnma$ ). The increase of  $x$  causes a decrease in the spin-coupling interaction between the moments of Mn. Hence, the ferromagnetic transition temperature decreases from 331 to 176 K. The saturated magnetization decreases from 80.36 to 78.33 emu/g due to the canted spin of the Sm moments.

*Key words:* mixed-valence manganite; rhombohedral; orthorhombic; spin-coupling

---

Due to the discovery of colossal magnetoresistance (CMR), there has been a renewed interest in the hole-doped magnetic perovskite compositions  $\text{Ln}_{1-x}\text{A}_x\text{MnO}_3$  ( $\text{Ln}=\text{La}, \text{Nd}, \text{Pr}$  and  $\text{A}=\text{Ca}, \text{Sr}, \text{Ba}, \text{Pb}$ ) with a  $\text{Mn}^{3+}/\text{Mn}^{4+}$  mixed valence [1,2]. A few possible mechanisms have been proposed for the magnetic phenomenon. Double-exchange (DE) interaction was proposed for the phenomenon based on the magnetic coupling between neighboring  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions that results from the motion of an  $e_g$  electron between two partially filled  $d$  shells with strong on-site Hund's coupling [3,4]. Recent reports showed that the mechanism of magnetic polarons formed by the Jahn-Teller distortion of the  $\text{MnO}_6$  octahedra [5]. The motion of the  $e_g$  electron can be strongly influenced by the average radius of the A site which exhibits a close relationship between the bond length and bond angle of  $\text{Mn}^{3+}-\text{O}^{2-}-\text{Mn}^{4+}$ . Previous research showed the fine tuning of magnetic properties could be achieved by the substitution of appropriate size of ions onto the

La-site [6]. In this work, the authors intend to study the substitution effect of La by smaller ion Sm in the  $\text{La}_{0.7-x}\text{Sm}_x\text{Pb}_{0.3}\text{MnO}_3$ .

Specimens of polycrystalline  $\text{La}_{0.7-x}\text{Sm}_x\text{Pb}_{0.3}\text{MnO}_3$  ( $0.0 \leq x \leq 0.3$ ) were synthesized by conventional solid-state reaction method. The structure and phase purity of the samples were examined by powder  $x$ -ray diffraction using  $\text{Cu-K}\alpha$  radiation at room temperature. The magnetization measurements at an 5 T applied field were performed by a Quantum Design MPMSR2-5S SQUID magnetometer. The zero-field-cooling (ZFC) and field-cooling (FC) at 5 K and 100 Oe were also obtained.

The  $x=0$  phase is indexed in a rhombohedral structure ( $R\bar{3}c$ ) while the rest exhibit orthorhombic symmetry ( $Pnma$ ) as illustrated in Fig.1. All the specimens were single phase with no detectable secondary phases. Figure 2 shows the temperature dependence of magnetization at 5 T for all compositions. The saturated magnetizations ( $M_S$ ), obtained from the saturated value of the magnetizing curve at 5 K and 5 T, decrease monotonically with increasing Sm content and tolerance factor  $t$  as shown in Table 1. The tran-

---

<sup>1</sup> Corresponding author. Present address: Department of Electrical Engineering, Hsiuping Institute of Technology, 11 Gunzeye Rd., Taichung 412, Taiwan. E-mail: hzc@mail.hit.edu.tw

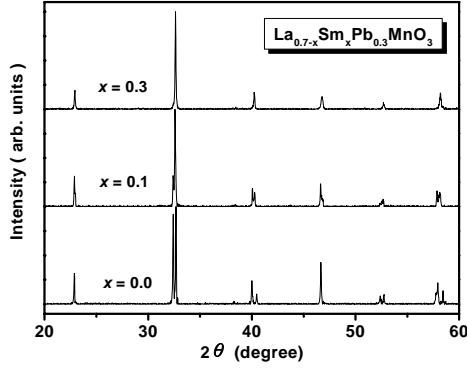


Fig. 1.  $x$ -ray diffraction patterns for all compositions.

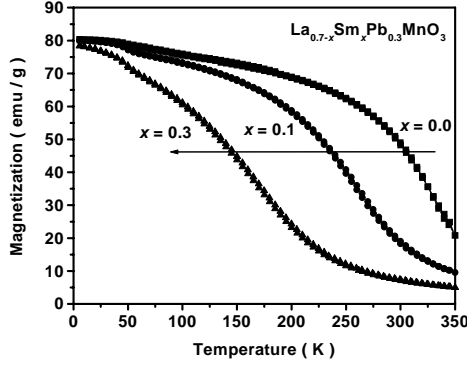


Fig. 2. The temperature dependence of magnetization at an applied field of 5T for all compositions. Arrow is guide for the eyes.

sition temperatures ( $T_C$ ), defined as the temperature where the value of  $dM(T)/dT$  reaches the maximum value, decrease as Sm content increases.

The ZFC-FC magnetization curves (see Fig.3) were measured at a field of 100 Oe in order to exam the spin order and magnetic behavior. 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. However, the irreversibility between the ZFC and FC magnetization curves is progressively seen as Sm content increases. The ZFC-FC curves display the irreversibility and  $\lambda$ -shape traces, indicating the tendency of a short-range spin ordering.

The transition of these results are in good agreement with the progressive substitution of small  $\text{Sm}^{3+}$  ( $1.24\text{\AA}$ ) for  $\text{La}^{3+}$  ( $1.36\text{\AA}$ ). The observation of the decrease of  $M_S$  and  $T_C$  when decreasing  $t$  means the deformation of  $\text{MnO}_6$ , the bending of  $\text{Mn-O-Mn}$ , the cant of Mn spins and the increase of competition between ferromagnetic DE and antiferromagnetic superexchange interaction. Therefore, from the results of this work, it is reasonable to conclude that the physical properties, such as crystallographic symmetry,  $M_S$

Table 1

Values of tolerance factor  $t$ , ferromagnetic transition temperature  $T_C$  and saturation magnetization moment  $M_S$ .

Composition	$t$	$T_C$ (K)	$M_S$ ( $\mu_B$ )
$\text{La}_{0.7}\text{Pb}_{0.3}\text{MnO}_3$	0.9915	331	80.36
$\text{La}_{0.6}\text{Sm}_{0.1}\text{Pb}_{0.3}\text{MnO}_3$	0.9872	264	80.19
$\text{La}_{0.4}\text{Sm}_{0.3}\text{Pb}_{0.3}\text{MnO}_3$	0.9785	176	78.33

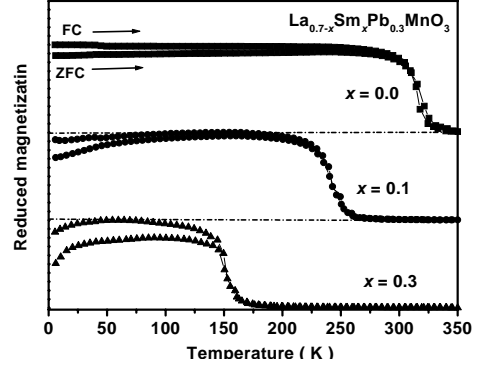


Fig. 3. The temperature dependence magnetization measurements at a low field of 100 Oe.

and  $T_C$ , are strongly depend on the mean size of La-site ions.

In summary, we have shown the transition of crystallographic and magnetic properties of the  $\text{La}_{0.7-x}\text{Sm}_x\text{Pb}_{0.3}\text{MnO}_3$  compounds. All these physical properties are controlled by the ionic size mismatch on the La-site and corresponding  $\text{Mn-O-Mn}$  bonds bending. Therefore, the saturated magnetic moment  $M_S$  increased, transition temperature  $T_C$  decreased and long-range spin order abated to short-range spin order can be seen induced by the substitution of La by Sm.

## Acknowledgements

This work was sponsored by the National Science Council of the Republic of China under the grant No. NSC 90-2112-M-164-001.

## References

- [1] J. M. D. Coey, M. Viret, *Advances in Phys.* **48** (1999) 167.
- [2] S. L. Young, *J. Appl. Phys.* **91** (2002) 8915.
- [3] C. Zener, *Phys. Rev.* **82** (1951) 403.
- [4] N. Shannon *et al.*, *Phys. Rev. B* **65** (2002) 104418.
- [5] A. J. Millis *et al.*, *Phys. Rev. Lett.* **77** (1996) 175.
- [6] J. P. Zhou *et al.*, *Appl. Phys. Lett.*, **75** (1999) 1146.