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

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Abstract

The magnetic and transport properties of mixed-valence manganites La$_{0.7-x}$Sm$_x$Pb$_{0.3}$MnO$_3$ ($x=0, 0.1$ and $0.3$) are investigated. The $x=0$ phase is indexed in a rhombohedral structure (R$3c$) 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 Ln$_{1-x}$A$_x$MnO$_3$ (Ln=La, Nd, Pr and A=Ca, Sr, Ba, Pb) with a Mn$^{3+}$/Mn$^{4+}$ mixed valance [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 Mn$^{3+}$ and 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 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 Mn$^{3+}$-O$^{2-}$-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 La$_{0.7-x}$Sm$_x$Pb$_{0.3}$MnO$_3$.

Specimens of polycrystalline La$_{0.7-x}$Sm$_x$Pb$_{0.3}$MnO$_3$ ($0.0\leq x\leq0.3$) were synthesized by conventional solid-state reaction method. The structure and phase purity of the samples were examined by powder x-ray diffraction using Cu-K$_\alpha$ radiation at room temperature. The magnetization measurements at an $5$ T applied field were performed by a Quantum Design MPMS2-5S 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$3c$) 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-
transition 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 examine the spin order and magnetic behavior. This is one of the methods 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 Sm$^{3+}$ (1.24 Å) for La$^{3+}$ (1.36 Å). The observation of the decrease of $M_S$ and $T_C$ when decreasing $x$ means the deformation of MnO$_6$, the bending of Mn–O–Mn, the cant of Mn spins and the increase of competition between ferromagnetic DE and antiferromagnetic super-exchange interaction. Therefore, from the results of this work, it is reasonable to conclude that the physical properties, such as crystallographic symmetry, $M_S$ and $T_C$, are strongly dependent on the mean size of La-site ions.

In summary, we have shown the transition of crystallographic and magnetic properties of the La$_{0.7-x}$Sm$_x$Pb$_{0.3}$MnO$_3$ compounds. All these physical properties are controlled by the ionic size mismatch on the La-site and corresponding 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.

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References