



Communication

Magnetocaloric effect and critical behavior in $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ ($x = 0.2, 0.4, 0.5$) manganitesM. Zarifi^a, P. Kameli^{a,*}, M. Mansouri^b, H. Ahmadvand^a, H. Salamati^a^a Department of Physics, Isfahan University of Technology, Isfahan 84156-83111, Iran^b Laboratoire de Physique des Matériaux, Faculté des Sciences de Sfax, Sfax University, B.P. 1171, 3000 Sfax, Tunisia

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ABSTRACT

This paper reports on an investigation carried out to determine the effects of Pr substitution on the magnetocaloric and critical behavior of $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ ($x = 0.2, 0.4, 0.5$) manganites. Structural studies show that all the compositions are single phase with a rhombohedral structure. Moreover, the lattice parameters and unit cell volume decrease with increasing Pr concentration while the Curie temperature (T_C) decreases with Pr^{3+} substitution for La^{3+} ions. Arrot's plots are used to determine the nature of the second order magnetic transition; no obvious hysteresis loss is observed in the vicinity of T_C in any of the samples. An exhaustive study of the critical behavior of the paramagnetic to ferromagnetic transition is conducted to determine the values of the critical exponents (β , γ , and δ) based on the modified Arrot's plot and using the Kouvel-Fisher method. We have obtained the critical exponents used 3D-Heisenberg and 3D-Ising models. It was found that with increasing Pr doping the values of β and γ decrease and increase, respectively. This suggests an increase in the ferromagnetic short-range order as a result of increasing Pr content in the $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ samples. Maximum changes in the magnetic entropy were found to be 4.16, 3.88, and $3.8 \text{ J kg}^{-1} \text{ K}^{-1}$ under an applied magnetic field of 5 T while the Relative Cooling Power (RCP) values reached 174.7, 252.3, and 255 J kg^{-1} for $x = 0.2, 0.4, \text{ and } 0.5$, respectively.

1. Introduction

Over the past few years, the perovskite manganites, with the general formula $\text{RE}_{1-x}\text{AE}_x\text{MnO}_3$ (RE: La, Pr, Nd, Sm, etc, and AE: Sr, Ca Ba, and Pb), have attracted much attention due to their potential technological applications [1,2]. Studies of manganites have revealed the potential magnetic refrigerating capability of these materials around the ferromagnetic-paramagnetic (FM-PM) phase transition [3,4]. Compared to conventional refrigerators, magnetic refrigerating materials enjoy such advantages as higher cooling efficiency, safer and environmentally-friendly nature, and convenience in miniaturization [5–8]. The working principle of magnetic refrigeration is based on the magnetocaloric effect (MCE) under a moderate applied field, which is characterized by Magnetic Entropy Change (ΔS_M) and Relative Cooling Power (RCP) [9,10]. In materials with MCE properties, first and second order transitions are accompanied by large variations in magnetization [11]. This warrants an investigation into the magnetization treatment at the phase transition temperature, which might be modified by the chemical composition.

In manganites, the partial replacement of trivalent rare-earth ions

of different sizes into the structure may bring about changes in the average radii of the ions. These changes, in turn, lead to disorders in the compound, which are believed to play the key role in controlling/modifying the values of ΔS_M and RCP [12]. Manganites with a first order magnetic phase transition (FOMT) exhibit high values of ΔS_M with a narrow ΔS_M curve and a large hysteresis, which lead to low operation frequencies and cooling power [13]. Therefore, the manganites with second-order magnetic phase transition (SOMT) are more suitable for magnetic refrigeration. A better understanding of the complex second order phase transition phenomena might be obtained by employing theoretical models to obtain the critical exponents.

The present work investigates the effects of Pr substitution on the critical exponents and magnetocaloric properties of $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ manganites through magnetization measurements. It is meant to build on the knowledge gained in previous studies of the $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ structure [14–19]. Pe, kala and Drozd [14,15] investigated the effects of grain size on polycrystalline $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ manganite. They reported that the transition from a polycrystalline structure to a nanocrystalline structure reduced the value of MCE and markedly broadened the temperature interval of MCE in structurally inhomoge-

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neous nanocrystalline samples. Amano et al. [16] reported on the effects of Ag^+ and Sr^{2+} cations on the MCE of $\text{La}_{0.8}(\text{Ag}_{1-x}\text{Sr}_x)_{0.2}\text{MnO}_3$ manganites and obtained a maximum $|\Delta S_M|$ value of $4.6 \text{ J kg}^{-1} \text{ K}^{-1}$ at $\mu_0 H = 5.0 \text{ T}$ in the sample with $x = 0.25$ and a relatively large refrigerant capacity (309 J kg^{-1}). Other researchers have also reported on Pr doping effects on manganites [20–24]. These studies show that the long-range FM order of the unsubstituted compound is destroyed as a result of Pr doping. Barik et al. [22] investigated the magnetocaloric effect in $\text{La}_{0.7-x}\text{Pr}_x\text{Ca}_{0.3}\text{MnO}_3$ compounds and reported a large MCE value and a very large RCP value for the compounds. They also observed that these values would decrease with increasing Pr content. Mleiki et al. reported that ΔS_M would decrease and that it would spread over a wider temperature range in their $\text{Sm}_{0.55-x}\text{Pr}_x\text{Sr}_{0.45}\text{MnO}_3$ samples with increasing Pr concentration [21].

The novelty of the present work lies in its investigation of the magnetic entropy of $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ ($x = 0.2, 0.4$, and 0.5), not hitherto reported elsewhere. In addition, the relationship between the critical exponent and doping is investigated for the first time.

2. Experimental

Polycrystalline samples of $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ ($x = 0, 0.2, 0.4, 0.5$) were synthesized using the standard solid-state reaction method. Briefly, the respective metal oxides and carbonates (i.e., La_2O_3 , MnO_2 , SrCO_3 , and Pr_6O_{11} with purities up to 99.9%) were mixed in the desired proportion. La_2O_3 was annealed in ambient air at 700°C for 7 h before use. After milling for 1 h, the mixture was calcined at $900, 1000$, and 1200°C for 16, 12, and 12 h, respectively. Finally, the obtained powder was pelletized and sintered at 1350°C for 24 h.

Phase purity and homogeneity were determined by powder X-ray Diffraction (XRD) at room temperature using a Philips EXPERT and a Cu-K α radiation source ($\lambda = 1.5406 \text{ \AA}$) in the range of $2\theta = 20 - 80^\circ$. Magnetization measurements versus temperature were carried out using a vibrating sample magnetometer (VSM). The isothermal magnetic entropy change (ΔS_M) was calculated from the magnetization versus applied magnetic fields up to 5 T at various temperatures.

3. Result and discussion

3.1. Structural characterization

Fig. 1(a) shows the XRD patterns of $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ ($x = 0, 0.2, 0.4, 0.5$) manganites. All the samples are single phase in nature without any detectable impurities within the measurement range studied. The XRD patterns were fitted by using the FULLPROF code. A typical XRD pattern of the sample with $x = 0.4$ along with its calculated pattern is presented in Fig. 1(b). Data analysis revealed that all the samples had a rhombohedral crystal structure with an $R\bar{3}C$ space group. The values of the "a" and "c" lattice parameters as well as the unit cell volume (V) are reported in Table 1 for all the samples. It is clear that the lattice parameters and unit cell volume decreased as a result of Pr doping due to the smaller average radii of the Pr (1.29 \AA) ions replacing at the La-site (1.34 \AA) of the perovskite structure.

3.2. Magnetic properties

Fig. 2 shows the field-cooled magnetization versus temperature, $M(T)$, of the samples exposed to a magnetic field of 1 kOe. A clear transition from PM to FM is observed. The transition temperatures estimated from these measurements are reported in Table 1. The Table 1 shows that the values for T_C decrease with increasing Pr concentration. The monotonic decrease observed in T_C and saturation magnetization can be explained by the decreasing average ionic radii of the A-site cations and the increasing cationic disorder of the site [25]. This certainly compresses the Mn–O–Mn bond angle but increases its length, leading to the reduced overlap between O-2p and Mn-3d

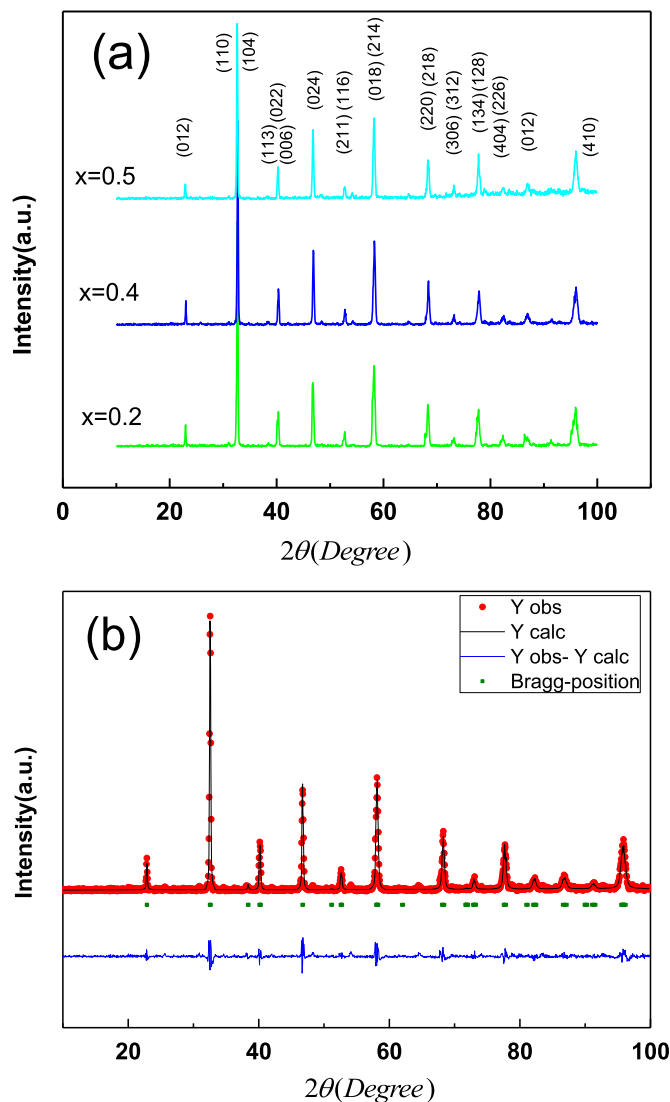


Fig. 1. (a) XRD patterns for $x = 0.2, 0.4$ and 0.5 samples at room temperature and (b) The experimental and calculated pattern of sample $x = 0.4$.

Table 1

Some obtained results for $\text{La}_{0.8-x}\text{Pr}_x\text{Sr}_{0.2}\text{MnO}_3$ compounds with $x = 0.2, 0.4$ and 0.5 .

Samples	$x = 0.2$	$x = 0.4$	$x = 0.5$
a (\AA)	5.5033	5.5049	5.4995
b (\AA)	13.4765	13.4388	13.4347
V (\AA^3)	353.4654	352.6892	351.8859
$\langle r_A \rangle$ (\AA)	1.2274	1.2201	1.2163
$\sigma^2 \cdot 10^{-4}$ (\AA^2)	19.1104	22.9800	24.5161
T_C	244	203	188
θ_p	270	241	225
μ_{eff}	5.69	6.20	6.37

orbitals which affect the double exchange interaction [26,27]. In addition, this could either be due to an imperfect antiparallel alignment between Pr^{3+} and Mn^{3+} moments, or it can be attributed to a hard alignment of Mn moments that originate from lattice distortions [28]. The temperature dependent inverse dc magnetic susceptibility ($\chi^{-1}(T)$) under $H = 1 \text{ kOe}$ is shown in the inset of Fig. 2(a) for all samples. Obviously, the $\chi^{-1}(T)$ curves exhibit a linear behavior at temperatures above T_C in the PM region; thus, all the samples obey the Curie–Weiss law which states that $\chi = C/(T-\theta_p)$, where C is the Curie constant and θ_p is the Weiss temperature. The linear fits of the samples also indicate that the evolution of FM interactions grows stronger with decreasing

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