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Room temperature critical behavior and magnetocaloric properties of $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}V_{0.1}O_3$

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Abstract

The magnetocaloric effect along magnetic phase transition and critical exponent in mixed manganite $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}O_3$ was investigated by dc magnetization measurements. Magnetic data indicate that the compound exhibit a continuous (second-order) paramagnetic (PM) to ferromagnetic (FM) phase transition. From the derived values of the critical exponents (β =0.385(1) and γ =1.481(3)), we conclude that $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}V_{0.1}O_3$ belongs to the three-dimensional Heisenberg class with short-range interaction. The maximum magnetic entropy ($-\Delta S_M$) and the relative cooling power (RCP) were found to be respectively, 4.266 J/kg K and 205.35 J/kg for a 5 T field change, making of this material a promising candidate for magnetic refrigeration near room temperature. From the field dependence of RCP and ΔS_M , it was possible to evaluate the critical exponents of the magnetic phase transitions. Their values are in good agreement with those obtained from the critical exponents using a modified Arrott method.

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Keywords: Manganites; Magnetocaloric effect; Relative cooling power; Critical exposents

1. Introduction

Recently, the perovskite manganites $\text{Re}_{1-x}\text{T}_x\text{MnO}_3$ (Re^{3+} = La^{3+} , Pr^{3+} , Nd^{3+} , etc.; T^{2+} = Ca^{2+} , Ba^{2+} , Sr^{2+} , etc.; ABO_3 type) had attracted considerable interest because they exhibit interesting physical effects and had potential applications due to the complex relationship between crystal structure, electrical, magnetic, and thermal properties, for example, the negative colossal magnetoresitance effect (CMR) and the magnetocaloric effect, the latter generally accompanied by a paramgneticferromagnetic transition [1-3]. One of the effective methods is to study in detail the critical exponents associated with the transition. In an earlier theoretical, critical behavior related to the paramgnetic (PM) to ferromagnetic (FM) transition of manganite in double exchange model was described within the framework of long range mean-field theory. However, the recent theoretical calculations have predicted the critical exponents in manganites in agreement with the short-range exchange interaction model [4,5]. Critical exponents for manganites show wide variation which covers almost all the universality classes even for the systems, when different experimental tools are used to determine them. So the interactions around the critical point will follow the scaling relations with critical exponents belonging to the conventional universality classes [6,7]. At present, the 3D Heisenberg model is extensively used to discuss critical properties and to understand short-range interaction in doped manganites [8]. A few experimental studies of critical phenomena have been previously made on same hole-doped as well as electron doped manganites [9,10]. It has been recently shown that there exists a universal curve for the magnetic entropy change for second order transition materials [11]. It can be constructed using a phenomenological procedure which does not require the knowledge of either the equation of state or the critical exponents of the material. Expressing the field dependence as $\Delta S_{\rm M}$ vs. H^n , this approach allows us to find a relationship between the exponent n and the critical exponents of the material and to propose aphenomenological universal curve for the field dependence of $\Delta S_{\rm M}$, which was successfully tested for series of soft magnetic amorphous alloys and lanthanide based crystalline materials. Up to now very little attention has been paid to the field dependence of RCP [12].

 $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}O_3$ is one of the extensively studied manganites which undergoes a paramagnetic metal to a

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Fig. 1. X-ray diffraction pattern and the corresponding Rietveld refinement of the $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}V_{0.1}O_3$ sample.

ferromagnetic metal transition around $T_{\rm C}=325$ K and it shows a $\Delta S_{\rm M}$ of 1.49 J kg⁻¹ K⁻¹ under $\mu_0 H=1$ T aroud its $T_{\rm C}$ [13].

As a contribution to the investigation of manganite materials, we report here the study of the effect, on the structural, magnetic, critical phenomenal and magnetocaloric properties, of V-doping in the $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}V_{0.1}O_3$ polycrystalline sample.

2. Experimental details

A polycrystalline sample $AMn_{0.9}V_{0.1}O_3$ (A=La_{0.6}Nd_{0.1} (CaSr)_{0.3}) compound were formed by a standard solid state reaction, used commercial powders (>99.99% purity) La₂O₃, Nd₂O₃, CaCO₃, SrCO₃, V₂O₃ and MnO₂ precursors. The detailed experimental process has been reported elsewhere [14]. The structure and phase purity of the prepared samples were checked by X-ray diffraction (XRD), using Cu-K α_1 radiation at room temperature. Magnetization vs. temperature and magnetic field curves were measured by using a Foner magnetometer equipped with a superconducting coil.

3. Results and discussion

3.1. Structural properties

Fig. 1 shows the XRD pattern after structural refinement using FULLPROF program [15], for the sample. The shifted Chebyshev polynomial with 10 variables was used to fit the background refinement and a Pseudo–Voigt function was selected to refine the shape of the peak. The analysis confirms that the structure of the compound was rhombohedral with R $\overline{3}$ c space group at room temperature and the stoichimetric nature of the sample is confirmed from the analysis. The refined lattice parametrs are a=5.412(2) Å, c=13.298(4) Å and V=347.55(2) Å³.



Fig. 2. Temperature dependence of magnetization under a magnetic field of 500 Oe for $La_{0.6}Nd_{0.1}(CaSr)_{0.3}Mn_{0.9}V_{0.1}O_3$ sample. The inset shows the temperature derivative dM/dT with T_C =298 K.

3.2. Magnetic properties

The investigation of the magnetic properties measured in a magnetic field of 500 Oe proved that the sample $AMn_{0.9}V_{0.1}O_3$ exhibit a single magnetic transition and behave in a ferromagnetic manner at a low temperature ($T \le T_C$) and in paramagnetic manner above the Curie temperature T_C ($T \ge T_C$) as shown in Fig. 2. This result confirms well the good quality of our sample. The temperature derivative of the *M*–*T* curve is shown in the inset of Fig. 2, from which the Curie temperature T_C ($T_C = 298$ K) can be univocally determined. This value is very close to the room temperature, which is quite suitable for magnetic refrigeration.

The magnetization M as a function of the applied magnetic field, at various temperatures, is shown in Fig. 3. Characteristic M(H) curves of manganites usually exhibit a very high increase in M at low fields and then a gradual saturation at high fields. The present sample also shows a steep rise in the low field range, the magnetization still increases steadily with increasing magnetic field and does not show any sign of saturation, even at 5 T. To analyze the nature of the magnetic phase transition in detail, we have carried out critical exponent study near $T_{\rm C}$ for AMn_{0.9}V_{0.1}O₃ sample.

3.3. Critical behavior

According to the scaling hypothesis [16], for a second order phase transition around the Curie point $T_{\rm C}$, critical exponents β (associated with the spontaneous magnetization $M_{\rm s}(H=0)$ below $T_{\rm C}$), γ (associated with the initial susceptibility $\chi = (\partial M/\partial H)$ above $T_{\rm C}$), and δ (associated with the critical isotherm $M(T_{\rm C}, H)$ at $T_{\rm C}$) are given as

$$M_{\rm S}(T) = M_{\rm o}(-t)^{\beta}, \quad t < 0,$$
 (1)

$$\chi_{o}^{-1}(T) = \left(\frac{h_{o}}{M_{o}}\right)t^{\gamma}, \quad t > 0$$
⁽²⁾

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