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Review

Effect of Cr substitution on magnetic and magnetic entropy change of $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{1-x}Cr_xO_3$ ($0.05 \le x \le 0.15$) rhombohedral nanocrystalline near room temperature

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ABSTRACT

We have studied the effect of Cr substitution on magnetic and magnetocaloric properties in nanocrystalline $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{1-x}Cr_xO_3$ ($x=0.05$, 0.1 and 0.15). The materials were prepared using the Pechini sol–gel method. All the studied samples were crystallized into a single phase rhombohedral structure with R−3C space group. Magnetic measurements indicate that the ferromagnetic double exchange interaction is weakened with increasing Cr concentration, resulting in a shift in T_C from 338 K to 278 K as x varied between 0.05 and 0.15. Detailed analyzes in the vicinity of the ferromagnetic (FM)–paramagnetic (PM) phase-transition temperature prove the samples undergoing a second-order phase transition. The magnetocaloric effect is calculated from the measurement of initial isothermal magnetization versus magnetic field at various temperatures. The maximum magnetic entropy change $|\Delta S_M^{max}|$ is found to decrease with increasing of Cr content from 4.04 J/Kg K for $x=0.05$–0.78 J/KgK for $x=0.15$ upon 5 T applied field change.

The relative cooling power (RCP) of $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{1-x}Cr_xO_3$ series is nearly 54% of pure Gd, which will be an interesting system for application in room temperature refrigeration.

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1. Introduction

In recent years, room-temperature magnetic refrigeration, based on the magnetocaloric effect (MCE), raised a great attention because of its potential advantages over the conventional vapor-cycle refrigeration [1–4]. Recently, many of the hole-doped manganites of the formula $R_{1-x}A_xMnO_3$ (R=La, Pr, etc. and A=Ca, Sr, etc.,) concerning second order magnetic transition, have been studied in an attempt to achieve a large MCE [5–8]. Furthmore, the manganites have additional advantages such as low cost, good chemical stability, easy preparation and more importantly the ability to control their magnetic transition temperatures (T_C) close to room temperature by R-site or Mn-site substitutions. One recent example of this chemical substitution for tuning T_C in manganites, is in the $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) system, which is a colossal magnetoresistive (CMR) ferromagnetic manganite with T_c ~370 K, and by substitution of the La ion by the rare-earths ions Eu, T_C is lowered [9].

Many theories have been proposed to understand the ferromagnetism and CMR of manganites such as double-exchange (DE) interaction, Jahn–Teller effect, and phase separation [10,11]. In hole-doped manganites, manganese ion exist in two different oxidation states, Mn^{3+} and Mn^{4+}, results in a Mn^{3+}/Mn^{4+} mixed-valence state, which creates mobile charge carriers and ferromagnetic ordering of Mn spins [12]. This behavior is usually interpreted with the help of DE mechanism, where the magnetic coupling between Mn^{3+} and Mn^{4+} ions results from the motion of an e_g electron between the two partially filled D-orbitals with strong on-site Hund's coupling [13]. Since the magnetic and MCE properties are dependent on the strength of DE interaction between Mn^{3+} and Mn^{4+} through oxygen atom, the doping at Mn-sites in manganites has a pronounced effect on the physical properties [14–16]. On the other hand, the ceramic preparation route, such as the conventional solid-state reaction method, the sol–gel method and combustion thermal spray techniques play an important role on the quality and the magnetocaloric properties of materials [17].

In this paper, we attempt to synthesize single phase, nanocrystalline $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{1-x}Cr_xO_3$ samples with $x=0.05$, 0.1, 0.15

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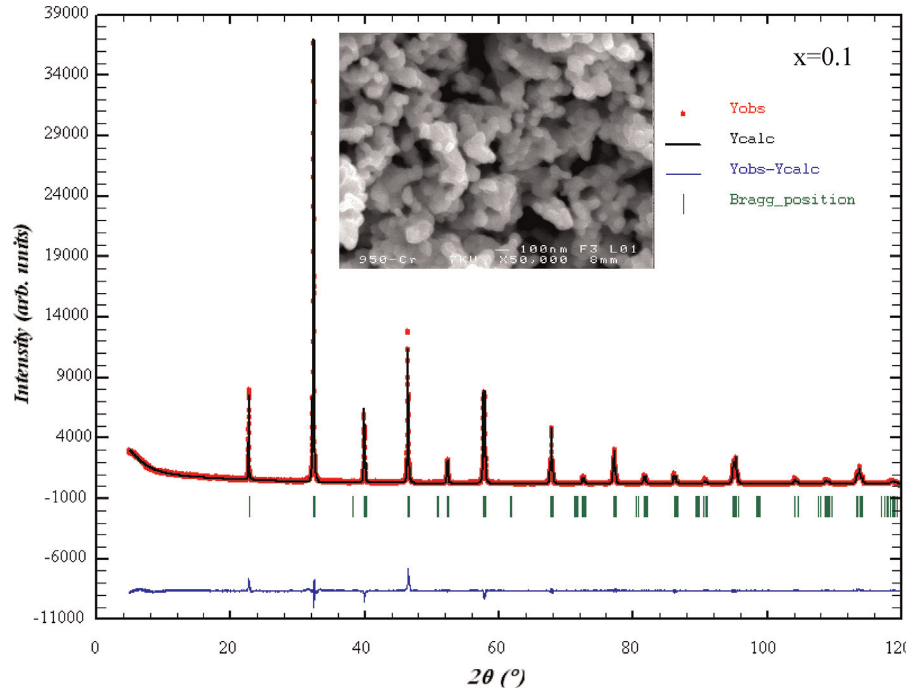

Fig. 1. Rietveld refinement profile for $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{0.9}Cr_{0.1}O_3$ sample performed using FULLPROF. Open circles correspond to experimental data and the lines are fits. Vertical bars are the Bragg reflections for the space group R−3C. The difference pattern between the observed data and fits is shown at the bottom. The inset represents a SEM micrograph for the sample.

Table 1
Refined structural parameters of $La_{0.65}Eu_{0.05}Sr_{0.3}Mn_{1-x}Cr_xO_3$ ($0.05 \le x \le 0.15$) samples at room temperature. Space group R−3C. *V* is the cell volume; B_{iso} is the overall isotropic thermal parameter, CS is the crystallite size evaluated from Rietveld refinements, R_{wp}, R_p and R_F are the agreement factors for the weighted profiles, the profiles and the structure factors; χ^2 is the goodness of fit. The numbers in parentheses are estimated standard deviations to the last significant digit.

Sample	*0.05*	*0.10*	*0.15*
Structure type	*Rhombohedral*	*Rhombohedral*	*Rhombohedral*
Space group	*R−3C*	*R−3C*	*R−3C*
Lattice parameter			
a(Å)	5.5144 (2)	5.5123(5)	5.5108 (2)
c(Å)	13.3779 (7)	13.3770 (1)	13.3704 (3)
V (Å^3)	352.182 (2)	352.011 (1)	351.653 (1)
$d_{(Mn,\ Cr)-O}$ (Å)	1.9574(7)	1.9563 (2)	1.9552(3)
$\theta_{(Mn,\ Cr-O-Mn,\ Cr)}$ (°)	166.97(3)	166.23 (3)	165.32(1)
W	0.09469	0.09480	0.09489
La/Eu/Sr			
x	0.000	0.000	0.000
y	0.000	0.000	0.000
z	0.25	0.25	0.25
B_{iso} (Å^2)	0.82(2)	0.75 (3)	0.59 (3)
Mn/Cr			
x	0.000	0.000	0.000
y	0.000	0.000	0.000
z	0.000	0.000	0.000
B_{iso} (Å^2)	0.61(4)	0.47(4)	0.34 (4)
O			
x	0.457(1)	0.456(4)	0.454(1)
y	0.000	0.000	0.000
z	0.25	0.25	0.25
B_{iso} (Å^2)	1.28 (3)	1.33 (5)	1.15 (2)
CS (nm)	74	68	63
Discrepancy factors (%)			
R_{wp} (%)	7.83	8.18	8.55
R_p (%)	6.02	6.28	7.34
R_F (%)	2.70	6.53	5.92
χ^2 (%)	2.32	3.47	3.48

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