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Review

Critical behavior and magnetic entropy change in the $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ perovskite

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ABSTRACT

Critical behavior in the $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ ceramics was studied using magnetization methods. Results show that the paramagnetic–ferromagnetic transition is of second order. Based on the critical behavior analysis using the Banerjee criterion and the Kouvel–Fisher method, we find the critical exponents: $\beta=0.395\pm0.010$, $\gamma=1.402\pm0.010$, and $\delta=5.208\pm0.007$, for which the magnetic interaction is satisfied within the three-dimensional Heisenberg model. Results indicate the presence of short-range interactions. The magnetic entropy change ($-\Delta S_M$) reached maximum values of 1.75, 1.45, 1.15, 0.8 and 0.43 J Kg^{-1} K^{-1} under a magnetic field variation of 5, 4, 3, 2 and 1 T, respectively. Nevertheless, these ($-\Delta S_M$) values are much low for any potential application at this moment. The nature of this phenomenon is discussed in relation to the characteristics of the magnetic phase transition and critical exponents.

1. Introduction

The magnetocaloric effect (MCE) consists of the variation of the magnetic entropy (ΔS_M) of a system in the presence or absence of a magnetic field in the vicinity of a magnetic transition. In general, compounds possessing a ferromagnetic order are the most interesting because they present an overall magnetic moment more important than compounds presenting a ferrimagnetic or antiferromagnetic order. In order to exhibit large MCE, materials should have large spontaneous magnetizations [1,2] as well as sharp drops of the magnetization with increasing temperature, associated to a ferromagnetic (FM)–paramagnetic (PM) transition at the Curie temperature [3–5].

The mean field theory has established direct relations between the magnetic entropy change and the magnetization, whereas the theory of critical phenomena justifies the existence of a universal magnetocaloric behavior in materials presenting second-order magnetic phase transitions [6,7]. Although lots of works have been done on the doping effects at the Mn-site, not much information is available about the magnetic transition itself, together with its associated parameters. Recently, it has been reported that 10 at% of Ga substituted on $La_{0.67}Ca_{0.33}MnO_3$ at the Mn site, can transform the first-order nature of the FM–PM transition into a second one, resulting in critical exponents close to the values predicted by the 3D (three-dimensional) Heisenberg model [8]. On the other hand, values reported so far in the literature concerning the critical exponents of various colossal magneto-resistance perovskite materials, have been found to be quite different to values corresponding to both long-range and short-range FM interactions. The critical behavior around the Curie temperature (T_C) has been studied previously by estimating the exponents β, γ, and δ, from the magnetization measurements. The values of these exponents may be close to those obtained by conventional models based on the mean-field theory, Ising or 3D Heisenberg models [9–11].

Therefore, in order to understand the nature of the magnetic transition in the $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ perovskite, we performed the investigation of its critical behavior at the FM–PM transition using dc magnetization data. We find that the critical exponents for $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ are close to those theoretically predicted by the 3D Heisenberg theory.

2. Experimental details

Polycrystalline $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ perovskite was synthesized by the conventional solid-state reaction method in air [12]. The X-ray powder diffraction (XRD) patterns were recorded with a "PANalytical X'Pert Pro" diffractometer with filtered (Ni filter) Cu radiation, in the range $20^\circ \leq 2\theta \leq 120^\circ$. The data were analyzed by the Rietveld method [12]. Magnetization (M) *versus* magnetic field ($\mu_0 H$), varying from 0 to 5 T at different temperatures (T), was measured using a MPMS-XL5 Quantum Design SQUID Susceptometer.

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3. Results and discussion

Fig. 1 shows the temperature dependence of the field-cooled (FC) and zero-field-cooled (ZFC) magnetizations for $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$. A clear FM–PM phase transition is observed corresponding to an increase of the magnetization (M) with decreasing temperature. At this step, the magnetic transition temperature T_C ($\sim$215 K) is defined as the inflection point of dM/dT (inset, Fig. 1). The ZFC and FC modes show an irreversibility between the two modes. As shown in our previous work for this sample [12], the cluster-glass component, if it does exist, is not the dominating mechanism.

3.1. Arrott plots

Fig. 2 shows the isothermal magnetization curves. In order to get a deeper insight on the type of the magnetic phase transition, the so-called Arrott plots of M^2-*versus*-H/M have been analyzed using the Banerjee criterion [13]. Results are presented in Fig. 3(a). According to this criterion, the magnetic transition is of second order if all the M^2-*versus*-H/M curves have positive

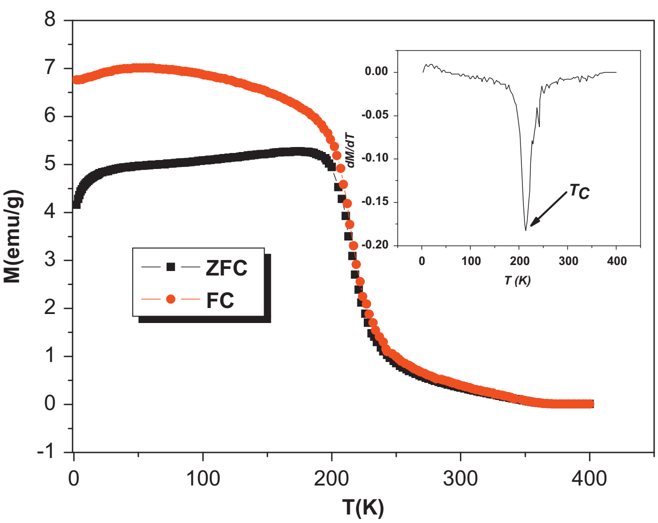

Fig. 1. Temperature dependence of the FC and ZFC magnetizations for $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$. The inset shows the temperature derivative, dM/dT *versus T*.

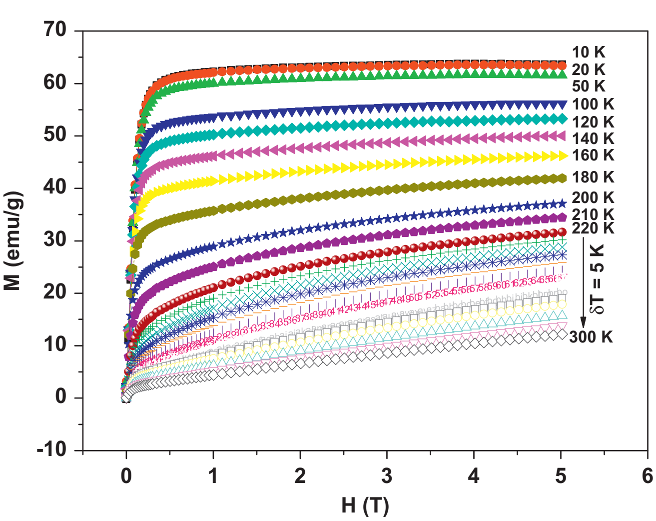

Fig. 2. Isothermal magnetization of $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ measured at given temperatures.

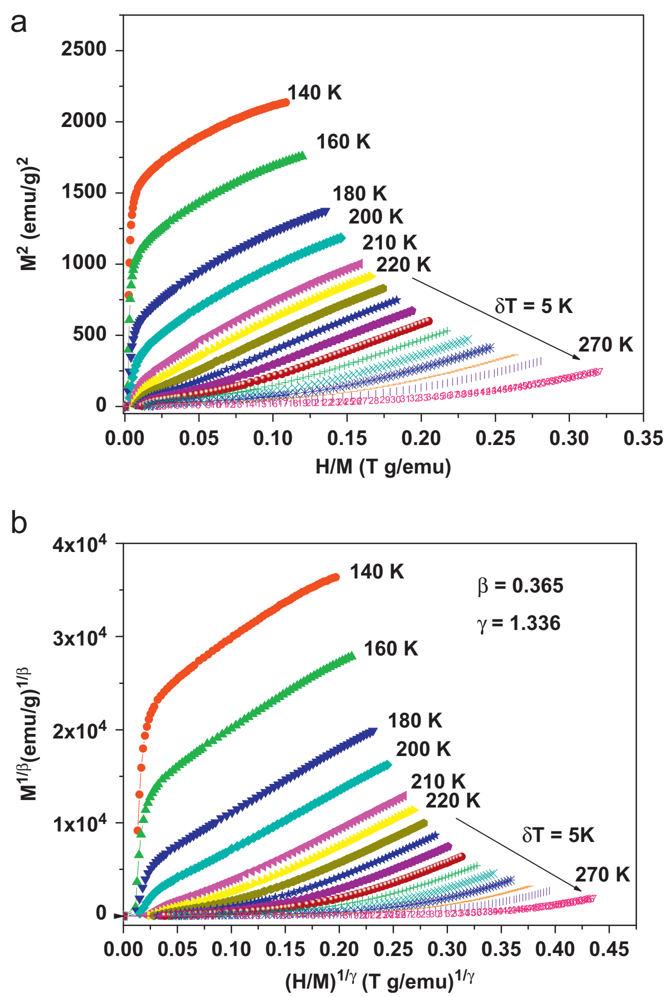

Fig. 3. (a) Standard Arrott plots (isotherms M^2 *versus* H/M). (b) Modified Arrott plots using 3D-Heisenberg model exponents.

slopes [14]. On the other hand, if some of the M^2-*versus*-H/M curves show a negative slope, the transition is of first order [14,15]. From Fig. 3(a) it can be observed that the M^2-*versus*-H/M curves show positive slopes, implying that these samples obey second-order magnetic transition mechanisms. The mean-field approximation can be generalized to the so-called modified Arrott plot (MAP) expression, based on the Arrott–Noakes equation of state [16]:

$$\left(\frac{H}{M}\right)^{1/\gamma} = \frac{T - T_C}{T_1} + \left(\frac{M}{M_1}\right)^{1/\beta} \quad (1)$$

where T_1 and M_1 are some constants which are characteristic of the material.

Mean-field theory values of $\beta = 0.5$ and $\gamma = 1$ [17] generate regular Arrott plots, characteristic of systems with long-range interactions (Fig. 3(a)). According to the mean field theory, the M^2-*versus*-H/M plots at various temperatures near T_C should show a series of parallel lines, and the line at $T = T_C$ should pass through the origin. In our case, the Arrott plots are not linear, implying that the mean-field theory is not valid for the $La_{0.6}Sr_{0.4}Mn_{0.8}Fe_{0.1}Cr_{0.1}O_3$ perovskite.

We have then analyzed our data by a modified Arrott plot method, based on the Arrott–Noakes equation of state (Eq. (1)). Fig. 3(b) shows the modified Arrott plots, $M^{1/\beta}$-*versus*-$(H/M)^{1/\gamma}$ using trial critical exponents β and γ for the 3D-Heisenberg model ($\beta = 0.365$, $\gamma = 1.336$ and $\delta = 4.8$) [17]. This choice produces a range