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Critical behavior studies in Ti-substituted lanthanum bismuth perovskite manganites

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ABSTRACT

Perovskite manganite $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xO_3$ (x = 0.05 and 0.1) synthesized using conventional solid state route method give rise to critical phenomenon in their magnetic interactions due to the substitution of non magnetic Ti ions. The critical behavior is observed near paramagnetic–ferromagnetic transition and is studied by magnetization measurements. Various techniques like Modified Arrott plot, Kouvel–Fisher method, scaling equation of state analysis and the critical magnetization isotherm were used to analyze the magnetization data on magnetic phase transition. The values of critical exponents β and γ obtained using different techniques are in good agreement. The obtained critical exponents are found to follow scaling equation with the magnetization data scaled into two different curves below and above the transition temperature, T_C. This confirms that the critical exponents and T_C are reasonably accurate. The obtained critical exponents for both the samples deviates from mean-field model and do not completely follow the static long range ferromagnetic ordering. This behavior is consistent with non magnetic nature of Ti substituted at Mn site and can be associated with Griffiths phase like phenomenon.

1. Introduction

Perovskite manganites, with general formula $La_{1-y}A_yMn_{1-x}M_xO_3$ (A = rare earth elements and M = transition elements) demand special attention because of the wide range of fascinating magnetic and transport properties involved with the interplay of charge, orbital, lattice and spin degrees of freedom [1]. Substituting the *A*-site cations with trivalent cations leads to dramatic change in their physical properties such as structural, magnetic and electrical transport properties. The trivalent cation such as Bi^{3+} at La^{3+} site has been extensively investigated due to its isovalence, whilst the mismatch of the A site cations caused by the substitution is small [2] and finds significant interest in the magneto-electronic (ME) coupling [3]. Furthermore considering the volatile nature of Bi^{3+} ions into account it is well understood that there exists a small amount of Mn^{4+} ions along with majority of Mn^{3+} ions [3]. The physical phenomena have been explained by effects such as 6 S^2 lone pair of Bi^{3+} [4], double exchange (DE) model [5], phase separation [6], Jahn–Teller (JT) distortion [7] and Griffiths phase [8]. The substitution of non-magnetic Ti ions at Mn site is also of particular interest from many decades due to the presence of additional effects of formation of significant instability in the Mn ion network [9]. After all the ionic radius of Ti^{4+} ion is found to lie in between the ionic radii of Mn^{4+} and Mn^{3+} which make the substitution interesting. Hence there exists a distinct possibility that a fraction of Ti^{4+} ions can occupy Mn^{3+} sites in addition to the Mn^{4+} sites. Recently it has been shown that though Ti^{4+} prefers to occupy Mn^{4+} sites, at high doping levels it also randomly distributed at Mn^{3+} sites leading to oxygen non stoichiometry (cation deficiency) or in-homogeneities [10,11]. Thus substitution of Ti should produce strain on Mn^{3+}–O–Mn^{4+} network by the formation of Mn^{3+}–O–Ti^{4+}chains. Similar kind of critical analysis for Ti substitution has been studied in $LaMn_{0.9}Ti_{0.1}O_3$ [12,13], $La_{0.7}Ca_{0.3}Mn_{0.9}Ti_{0.1}O_3$ [14] and $La_{0.57}Nd_{0.1}Pb_{0.33}Mn_{1-x}Ti_xO_3$ [15]. The substitution of Ti at Mn site gives rise to inhomogeneous short range interactions in the magnetic ordering around T_C. And the Ti substituted samples mentioned above studied by other researchers explicitly confirm this short range ordering which falls under 3D Heisenberg (HM) universality class [12–15]. Further

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Xuebin Zhu et al. [14] clearly observed that the low Ti substituted sample $La_{0.7}Ca_{0.3}Mn_{0.95}Ti_{0.05}O_3$ show the characteristics of long-range interactions and for the higher Ti substituted sample, $La_{0.7}Ca_{0.3}Mn_{0.9}Ti_{0.1}O_3$ gives the evidence of spin-glass like characteristics with short-range magnetic interactions [14]. Hence the fundamental magnetic characteristics in these manganites containing non-magnetic ions have not been clarified yet or fully understood.

In order to understand these effects, we have investigated the critical behavior and nature of magnetic interactions near phase transition temperature, T_C in $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xO_3$ (x = 0.05 and 0.1) perovskite manganite. The aforesaid sample is basically a ferromagnetic insulator having tetragonal perovskite structure which belongs to I4/mcm space group [11]. In our previous studies we have reported spin glass (SG)/cluster glass (CG) and Griffith's phase behavior at very low and intermediate temperatures in $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xO_3$ with x = 0.05 and 0.1 samples respectively [10,11]. Now in present work we focus on the critical behavior near the magnetic phase transition and their analysis using various techniques such as modified Arrott plot (MAP), Kouvel–Fisher (KF) method, scaling-equation-of-state analysis and the critical magnetization isotherm. The theoretical critical values of β, γ and δ associated with their respective magnetization exponents for ideal short range magnetic interaction as per 3D Heisenberg model (HM) must be equal to 0.365, 1.386 and 4.800 respectively [16]. And for long range interaction as per theoretical mean field model (MFM) must have ideal critical exponents of 0.5, 1.0 and 3.0 respectively [16]. The critical exponents β, γ and δ values obtained using above methods in this work are found to be in close agreement with different models and satisfy scaling equation of state, but do not indicate static long range FM ordering associated with MFM around the magnetic phase transition.

2. Experimental techniques

The detailed synthesis of polycrystalline samples of $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xO_3$ with $x = 0.05$ and 0.1 using conventional solid state reaction method [17] are already been reported [10,11]. The samples were characterized using X-ray diffraction (XRD), scanning electron microscope (SEM) equipped with energy dispersive X-ray spectroscopy (EDS) and the results obtained are already been discussed in detail [11]. DC magnetization as a function of temperature was carried at 500 Oe and for critical behavior study, DC magnetization measurements as a function of magnetic field ($0 \leq H \leq 5T$) at various constant temperatures in the range of 30 K $\leq T \leq$ 60 K at intervals of 2 K were carried out using Superconducting Quantum Interference Device-Magnetic Properties Measurements Systems (Quantum Design SQUID-MPMS XL 5).

3. Results and discussions

DC Magnetization as a function of temperature (M vs. T) measured in field cooled (FC) mode at constant magnetic field, H = 500 Oe for $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xMnO_3$ with x = 0.05 and 0.1 samples are shown in Fig. 1. At temperature 10 K, the saturation magnetization value for the sample with x = 0.1 is almost less than 50% of the saturation magnetization of sample with x = 0.05. The obtained results are consistent as the non-magnetic Ti ion decreases the magnetization in the sample. We have determined the paramagnetic (PM) to ferromagnetic (FM) transition, Curie temperature, T_C to be ~45 K for both x = 0.05 and 0.1, at which dM/dT reaches its minimum as shown in the inset of Fig. 1.

From our earlier study, the magnetic T_C of the parent compound $La_{0.4}Bi_{0.6}MnO_3$ was found to be 68 K [11]. A considerable decrease in T_C can be observed with Ti (x = 0.05, i.e. 5%) substitution at Mn site. The observed decrease in the magnetic T_C is in agreement with our earlier findings explained in the context of magnetic dilution [10,18]. However, with increasing the Ti substitution to x = 0.1 (i.e. 10%) T_C almost remained constant. The observed behavior is analogous with results of Cao et al. for $La_{0.7}Ca_{0.3}Mn_{1-x}Ti_xO_3$ sample (2–6 % of Ti doping) and has been explained due to pinning of Mn^{3+}/Mn^{4+} ions at a fixed value by the substitution of Ti^{4+} which limits the fluctuation of Mn Valances [9]. Although in the paper by Cao et al. [9], the reported values of T_C decreased beyond 8 and 10% substitutions of Ti concentration, but in our case the values of T_C decreased beyond 10% (i.e. for 50% of Ti substitution where T_C was found to be at 40 K [10,19]). This behavior can be due to different parent compound.

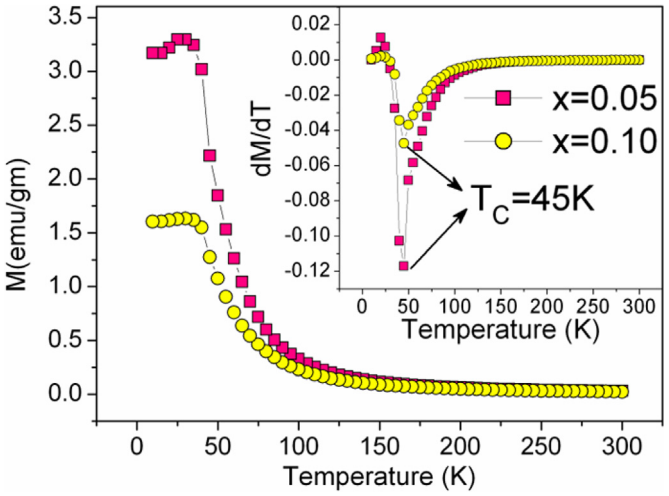

Fig. 1. Temperature dependence magnetization (emu/gm) in an applied field of 500 Oe are displayed for the synthesized $La_{0.4}Bi_{0.6}Mn_{1-x}Ti_xO_3$ with x = 0.05 & 0.1. Inset displays derivative of magnetization (dM/dT) for the respective samples.

A set of magnetization isotherms were measured around PM–FM like transition region for both $x = 0.05$ and 0.1. Fig. 2a and b shows the magnetization isotherms measured in the temperature range of 30 K $\leq T \leq$ 60 K at a temperature increment of $\Delta T \approx 2$ K. Inset of Fig. 2a and b we show M^2 vs. H/M plots over a wide range of temperature to determine the order of the magnetic phase transition. The above plots are based on criterion given by Banerjee et al. [20]. The criterion is used to inspect the slope from M^2 vs. H/M isotherm plots. The plots shown in inset of Fig. 2a and b indicates positive slopes in the complete M^2 range for both the samples, which confirms that the samples exhibit a second order FM–PM phase transition (absence of thermal hysteresis).

Also it is well observed that the curves are not parallel either to each other and deviates from straight line at high field regime. This infers that the magnetization of these samples does not obey ideal mean field approximation (β = 0.5, γ = 1). Moreover, in Fig. 2a and b the Arrott plots didn't exhibit spontaneous magnetization which is direct evidence to the absence of ideal long range static FM interactions in the studied samples. This particular behavior in some of the manganites was explained due to presence of Griffith's phase [21–23]. In general, the Griffiths singularities is characterized by deviation from Curie–Weiss (CW) linear law in paramagnetic region around T_C and the value of susceptibility exponent λ ($0 < \lambda \leq 1$) has been obtained from the relation $\chi^{-1} = (T - T_C^R)^{1-\lambda}$ [24–26]. In our earlier report [10] the studied samples explicitly showed deviation from standard Curie–Weiss (CW) linear law in paramagnetic region around T_C and obtained values of λ lie in between 0 and 1 (presented in Table 1). These observations suggested the existence of the Griffith's phase in the studied samples.