



Correlating melting and collapse of charge ordering with magnetic transitions in $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$



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ABSTRACT

The magnetic properties of polycrystalline $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$ material are investigated at different temperatures. The existence of magnetically diverse phases associated with various relaxation modes and their modulation with temperature and doping is analyzed. $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ exhibited field induced melting and collapse of charge ordered antiferromagnetic (CO-AFM) phase into ferromagnetic (FM) state. This phenomenon results in lowering of Neel's temperature (T_N) along with changes in the slope of magnetic moment with temperature. Using normalized $M(T)$ curves, the variation and interplay of charge ordered temperature (T_{CO}), Curie temperature (T_C) and T_N is conferred. The coexistence of two magnetic modes is explained as major ingredient for the magnetic transitions as well as metal to insulator transition (MIT); where melting and collapse of charge ordering is conversed as basic feature in these Praseodymium (Pr) doped $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ materials.

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1. Introduction

For more than a decade, the coexistence of different phases in mixed valence manganites ($\text{R}_{1-x}\text{A}_x\text{MnO}_3$; where R is a rare earth element and A is a divalent alkaline earth element) have attracted considerable attraction due to their exotic electrical, magnetic and structural properties [1,2]. The varied range of electronic and magnetic phases in these manganites is due to the competition of different interactions, mainly between double exchange and Jahn Teller interactions [3]. The appearance of metal-insulator transition in these types of strongly correlated electron systems is often characterized through physical complexity of the competition of different coexisting phases [4]. Short band width $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ (LCMO) and intermediate band width $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ (PCMO) oxides have been in focus for complex electronic and magnetic behaviors due to combination of lattice, charge, spin and orbital degree of freedom [5,6]. It is already known that electron mobility

and bandwidth depends more on the relative strength of localizing JahnTeller interactions as compared to the hopping energy [7]. Therefore, depending upon bandwidth, the charge ordered insulating phase (arranged in checkerboard symmetry) can be melted by applied field thereby destabilizing the charge exchange magnetic order in favor of ferromagnetic (FM) ordering. While, the melting and collapse of charge ordering (CO) under external perturbations such as magnetic field, pressure and current has been reported in the literature [4,8,9], the origin of these phenomena is still not understood clearly.

Depending upon the thermal and magnetic history, LCMO exhibits a competition between FM and antiferromagnetic (AFM) states [6,10] in both tetragonal as well as orthorhombic structures [11,12] with growth of nano phase regions below T_C [1]. LCMO is reported to exhibit change in crystal symmetry ($T_C = 225$ K) along with a transition from paramagnetic (PM) to ferromagnetic metal (FMM) and then further to a charge exchange type antiferromagnetic insulating (CE-AFI) phase below $T_N = 155$ K [6,12]. PCMO is among the most interesting materials due to its appealing electrical and magnetic properties showing charge and orbital ordering along with CE-AFI phases at $T_{CO} = 245$ K and $T_N = 155$ K,

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respectively [13,14]. In our recent report [15], a detailed alternating current (ac) electrical study of phase separated PCMO sample is carried out, where the presence of various electro active regions at different temperatures is established. Different conduction mechanisms and metal to insulator transition (MIT) at different temperatures is reported in an ac electrical study of phase separated $\text{La}_{0.50-x}\text{Pr}_x\text{Ca}_{0.50}\text{MnO}_3$ (LPCMO- x) materials [16]. Generally, the electrical properties are described on the basis of shortest path available between two electrodes while the interactions of the spin orientations present in the specific phases are addressed by magnetic studies [17]. This manuscript is intended to address the magnetic correlations of these samples with A-site cationic replacement. In similar type of materials, decrease in temperature can change the lattice parameters around charge ordering temperature [18] and suppress the charge ordered phase with appearance of weak ferromagnetism [19]. Although, the presence of CO and T_N in both compounds (LCMO and PCMO) is common, their interplay with ferromagnetic/antiferromagnetic domains is not clearly understood.

In order to understand the correlation of relaxation modes (of different electroactive regions) with the metal to insulator transition [16], it is anticipated that the mixing of these two compounds is the next logical step where interplay of two phases will dictate fascinating magnetic properties. Metal to insulator transition (MIT) is reported for $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$ (LPCMO- x) materials [16] and other similar types of manganite materials [20,21]. This manuscript will address the interactions relating to spin degree of freedom and magnetic correlations for these intermediate and short bandwidth mixed ceramics. $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$ ($x=0.00$ to 0.40) are analyzed for variations in magnetic properties at low temperatures. At A site, doping of divalent at trivalent cations results in an inhomogeneous distribution of Mn^{3+} and Mn^{4+} ; this amendment can alter Mn^{3+} -O- Mn^{4+} network linkages which can change both of the magnetic and electrical properties [22]. In the present work, $\text{Mn}^{3+}/\text{Mn}^{4+}$ ratio is assumed to be fixed and the change of A site cation is conferred to bring changes only in the bond angle of Mn^{3+} -O- Mn^{4+} along with alteration in the Mn-O bond length [23]. We probed the phase coexistence, melting and collapse of charge ordering phenomena arising from doping of Pr^{3+} at La^{3+} site in LCMO.

2. Methodology

Polycrystalline $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$ (LPCMO- x) samples are prepared by solid state reactions method. Stoichiometric amount of La_2O_3 , Pr_6O_{11} , Mn_2O_3 and CaCO_3 are thoroughly mixed and grinded in an electric grinder. Calcination is carried out at different temperatures from 1000°C to 1300°C for 10 h with intermediate grindings. After calcination these oxides are then pressed into cylindrical shape pellets of 8 mm diameter and 2 mm thickness by applying a uniaxial load of 0.4 tons for one minute. Further compactions of these pellets are carried out by applying an isostatic pressure of 240 MPa for two minutes using a cold isostatic press (CIP). These pellets are finally sintered in a furnace at 1350°C for 10 h with heating and cooling rates of $2^\circ\text{C}/\text{min}$ [15,16]. Synthesized powder is subjected to XRD analysis for phase identification. Magnetization measurements are carried out by a cryogen free Physical Property Measuring System (PPMS) of the Cryogenic Ltd., UK (installed at the central diagnostic laboratory, PINSTECH) equipped with $\pm 7\text{T}$ magnetic fields. Magnetic studies of LPCMO- x are conducted in zero field cooling mode under the modulation of magnetic field in the temperature range of 10 K to 300 K. Abbreviations used for the studied samples in the manuscript are; (LPCMO- x) for $\text{La}_{0.5-x}\text{Pr}_x\text{Ca}_{0.5}\text{MnO}_3$, (LCMO) for $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$, (LPCMO-2) for $\text{La}_{0.3}\text{Pr}_{0.2}\text{Ca}_{0.5}\text{MnO}_3$, (LPCMO-3) for $\text{La}_{0.2}\text{Pr}_{0.3}\text{Ca}_{0.5}\text{MnO}_3$ and (LPCMO-4) for $\text{La}_{0.1}\text{Pr}_{0.4}\text{Ca}_{0.5}\text{MnO}_3$.

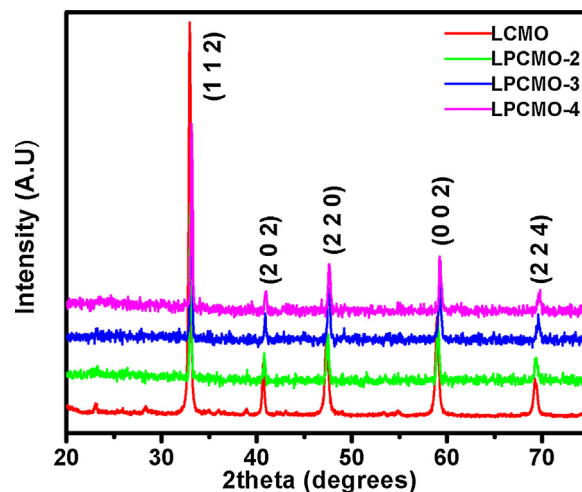


Fig. 1. X-ray diffraction patterns of LPCMO- x samples.

3. Result and discussions

Fig. 1 shows X-ray diffraction patterns of all LPCMO- x samples at room temperature. All peaks are indexed properly and there is no signature of any impurity in these LPCMO- x samples. With Pr doping all peaks shifted to higher 2θ values [16]. Fig. 2 shows the magnetization $M(T)$ plots against temperature at different applied magnetic fields. The characteristic variations in $M(T)$ with magnetic field is explained by the presence of different magnetic transition temperatures in the LCMO sample where increasing magnetic field and lowering temperatures reveal two distinct spectral features as shown in Fig. 2(a). One feature i.e., major peak is observed around 150 K which coincides with the reported Neel temperature (T_N) of LCMO sample [11]. Amplification of magnetic field results in shifting of T_N towards lower temperatures. The peak height is increased with magnetic field showing FM correlations. For field up to 0.5 T the magnitude of the moment showed small changes. However, for 3 T magnetic field moments approached to 85emu/g at $\approx 120\text{K}$. In view of above results, there are two possible arguments for this variation in the moment of LCMO sample. First opinion suggests that the FM alignment is competing with the antiferromagnetic one in this temperature range. Whereas, second point of view can be that the FM correlations are established in nano or small regions which grow under applied magnetic field [1]. At temperatures just below this major peak, a sharp decrease in the moment of LCMO is observed which is consistent with the literature for the occurrence of antiferromagnetic (AFM) transition [24]. The second feature in Fig. 2(a) is defined as minor peak around 210 K and coincides with the reported Curie temperature (T_C) of this material [25]. This feature is visible up to 1 T due to competition of charge-ordered AFM phase with the long range ferromagnetic order (hence termed T_{CO}). However, at the higher applied field, i.e., 3 T, this minor peak (T_{CO}) is faded away which is argued to manifest FM domains correlation around this temperature range with an increase in overall magnetic moment of the material [26].

In order to elaborate the variation of magnetic moment and magnetic transitions, the magnetic data shown in Fig. 2(a) is transformed into normalized magnetization with respect to magnetization at 300 K [13]. Fig. 2(b) shows variation of major peak, which is shifted to lower temperatures with magnetic field, although, the temperature of minor peak corresponding to T_C is not changed. These results can be summarized for the successive magnetic phases with lowering of temperature, namely, PM from room temperature to charge ordered state which is in competition

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