Specific heat studies on the charge and magnetic ordering in manganites

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Abstract

Specific heat (C) studies with a wide range of temperatures (T = 80-300 K) on polycrystalline manganites $Pr_{1-x}Ca_xMnO_3$ (113-PCMO) with x=0.3-0.4, $La_{1-y}Ca_yMnO_3$ (113-LCMO) with y=0.3 and 0.5, and $La_{2-2z}Sr_{1+2z}Mn_2O_7$ (327-LSMO) with z=0.3 and 0.5 are reported. Clear anomalies in C are identified which are associated with charge ordering (CO), antiferromagnetic (AFM) and ferromagnetic (FM) transitions. The anomalies in 113-PCMO and 113-LSMO are bigger than those of 327-LSMO. The possible explanations are discussed on the basis of dimensionality effects.

Key words: Specific Heat; Charge ordering ; Magnetic ordering ;Mangantites

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The manganites of the Ruddlesden-Popper series, $R_{n+1}Mn_nO_{3n+1}$ (R=trivalent rare earth metals and n accounts for the dimensionality) illustrate many interesting properties like colossal magnetoresistance (CMR), charge ordering (CO) and magnetic field induced transitions when R is partially substituted by a divalent cation A [1,2]. The temperature dependence of specific heat (C) of these manganites clearly indicates the anomalies corresponding to the various phase transitions [3]. The present paper deals with the comparative specific heat study of 3D infinite layered manganites (113-PSMO and 113-LSMO) and quasi-2D bi-layered 327-LSMO. Polycrystalline $Pr_{1-x}Ca_xMnO_3$ (113-PCMO) with x=0.3-0.4, $La_{1-y}Ca_yMnO_3$ (113-LCMO) with y=0.3 and 0.5, and $La_{2-2z}Sr_{1+2z}Mn_2O_7$ (327-LSMO) with z=0.3 and 0.5 oxides were synthesized by a standard solid-state-reaction method using the powder raw materials Pr_6O_{11}, La_2O_3, CaO, $SrCO_3$ and MnO_2 with purity 99.9 percent or better. For details of preparation, please see Refs. [4,5]. Relative specific heat measurements in high temperature (80-300K) were performed with a high resolution ac calorimeter using chopped light as the heat source [6].

Figure 1 shows the temperature variation of specific heat (C) of $Pr_{1-x}Ca_xMnO_3$ (113-PCMO) manganites. Clear anomalies for AFM ($T_N \sim$ 160-165K) and CO ($T_{CO} \sim$ 215-227K) transitions are observed in samples with x=0.35-0.4 and the transition temperatures increase with the increase of x. While x=0.3, the anomaly in C due to AFM or CO transition is smeared out. It is well established that the CO state is most stable with x=0.5 and is destabilized with the decrease of x. As the CO transition vanishes with x≤0.3, the boundary value of x for CO transition in this system is 0.3. Temperature dependence of C for $La_{1-y}Ca_yMnO_3$ (113-LCMO) with y=0.3 and 0.5, and $La_{2-2z}Sr_{1+2z}Mn_2O_7$ (327-LSMO) with z=0.3 and 0.5 samples are displayed in Fig. 2. The anomalies for FM transition ($T_C \sim$ 100K) in $La_{1.4}Sr_{1.6}Mn_2O_7$,

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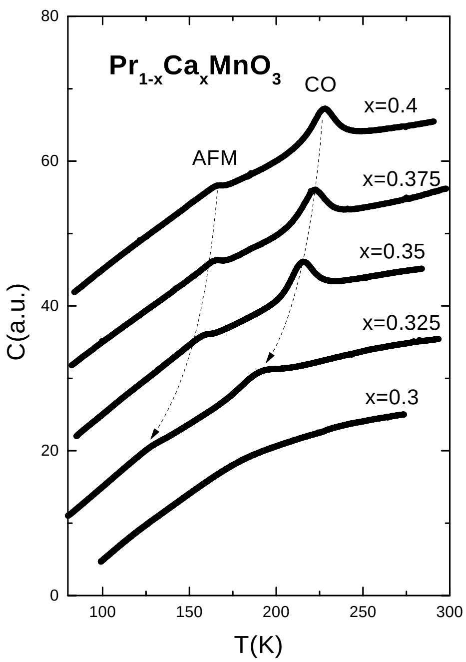

Fig. 1. Temperature variation of specific heat (C) of $Pr_{1-x}Ca_xMnO_3$ (113-PCMO) with x=0.3-0.4 manganites.

CO transition ($T_{CO} \sim 220$ K) in $LaSr_2Mn_2O_7$, FM transition ($T_C \sim 230$K) in $La_{0.7}Ca_{0.3}MnO_3$ and AFM ($T_N \sim 130$K) as well as CO ($T_{CO} \sim 180$K) transitions in $La_{0.5}Ca_{0.5}MnO_3$ can be clearly identified. The magnitude and temperature of specific-heat anomalies exhibited in 113-LCMO and 113-PCMO are consistent with those reported [3]. Interestingly the trivalent rare earth plays a dominant role in magnetic transitions as a very large peak is observed in $La_{1-x}Ca_xMnO_3$ for FM transition (Fig.2) where as no definite peak is detected in $Pr_{1-x}Ca_xMnO_3$ (Fig.1) for x=0.3 in the temperature range of present investigation.It also deserves noting that the magnitudes of specific heat anomalies observed in quasi-2D manganites (327-LCMO) are much smaller than those in 3D ones (113-PCMO). This may be due to the 40 percent higher density of Mn atoms (18.23 Mn atom/nm3) with infinite number of MnO_6 layers along the c direction in 3D systems compared to the quasi-2D one in which the density of Mn atoms is 13.18 Mn atom/nm3 and has double MnO_6 layers along the same direction. The 2D character of magnetism in bi-layered manganites is also caused by the reduction of exchange coupling along the c direction. Hence the stronger exchange interaction in 3D perovskites compared to 2D bi-layered systems may give rise to bigger specific heat anomaly in the former compared to the latter. This dimensionality effect on the entropy change associated with the thermodynamics of phase transitions in these manganites require further detailed quantitative analysis.

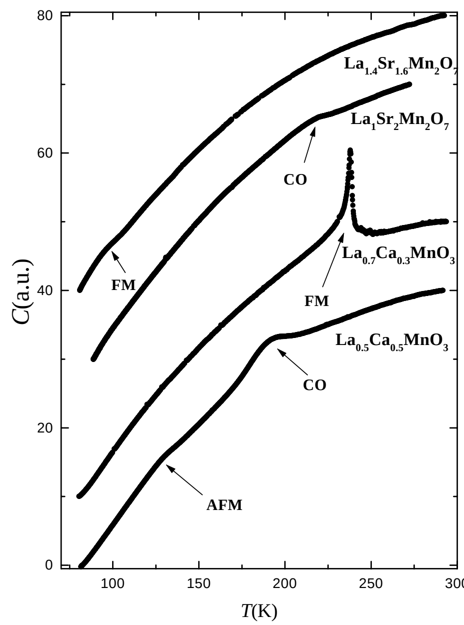

Fig. 2. Temperature dependence of specific heat (C) of $La_{1-y}Ca_yMnO_3$ (113-LCMO) with y=0.3 and 0.5, and $La_{2-2z}Sr_{1+2z}Mn_2O_7$ (327-LSMO) with z=0.3 and 0.5.

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