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Effect of Co doping on the magnetic properties of $\text{La}_{0.85}\text{Ag}_{0.15}(\text{Mn}_{1-y}\text{Co}_y)\text{O}_3$

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Abstract

The mixed valent manganites $(\text{La}_{0.85}\text{Ag}_{0.15})\text{MnO}_3$ with perovskite structure has been prepared by doping up to 50% of Co at the Mn site. Paramagnetic (PM) to ferromagnetic (FM) transitions have been observed in all the prepared materials. However, the long-range magnetic ordering observed in $(\text{La}_{0.85}\text{Ag}_{0.15})\text{MnO}_3$ is systemically reduced to cluster glass-type (short-range) of FM ordering due to the introduction of Co. The FM transition temperature was found to decrease with increase in Co doping up to 20% and for further increase in Co doping, the T_c was found to increase. They are explained on the basis of competition between FM double exchange interactions in Mn–O–Mn and Co–O–Co networks. In addition to PM–FM transition, evidences of FM to antiferromagnetic (AFM), and AFM to reentrant spin-glass transitions have been observed. The shift in spin-glass freezing temperature, T_f has been observed from the frequency variation of ac susceptibility measurements. The observed magnetic transitions are explained on the basis of magnetic interactions in different Mn–O–Mn, Mn–O–Co and Co–O–Co networks and such transitions are also observed from the measurement of third harmonic susceptibility. Metal–insulator transition and colossal magneto-resistivity have been observed up to 10% of Co doping.

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1. Introduction

The perovskite manganites, $\text{R}_{1-x}\text{A}_x\text{MnO}_3$ (R = rare earth elements, A = alkaline earth, alkali elements, etc.) are known to exhibit very interesting structural, electrical and magnetic properties depending on the doping concentration, x [1–4]. In addition to that, such interesting electrical and magnetic properties have been reported in cobalites $\text{La}_{1-x}\text{A}_x\text{CoO}_3$ [5,6]. There are several reports on the substitution of transition elements in place of Mn to study their interactions with Mn ions [7–16]. The doping of Co on Mn site has been studied by several groups, especially in $\text{La}_{0.7}\text{A}_{0.3}\text{MnO}_3$ compounds (A = Ca, Sr, Ba) [7–12]. The Co doping leads to reduction in ferromagnetic (FM) transition temperature, T_c , and introduction of competing magnetic interactions in Mn–O–Mn, Mn–O–Co

and Co–O–Co networks. The various low temperature anomalies observed in magnetization and susceptibility measurements of Co doped materials are yet to be understood completely. To investigate the different interactions between Co and Mn ions, we have prepared polycrystalline $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$ compounds. Reentrant spin-glass (RSG) like transition was studied from the frequency variation of ac susceptibility measurements. Multiple magnetic transitions such as paramagnetic–ferromagnetic (PM–FM), ferromagnetic–antiferromagnetic (FM–AFM) and low temperature RSG transition have been observed from the third harmonic ac susceptibility measurements.

2. Experimental techniques

The $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$ compounds were prepared for $y=0$ to 0.50 by solid state route. The final sintering in pellet form was carried out at 1000 °C for over 36 h. X-ray

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diffraction (XRD) patterns were recorded at room temperature using a Bruker D8 Advance XRD machine by employing CuK_α radiation. The average valency of Mn ions was determined by the chemical titration technique as described in Ref. [15]. DC electrical resistivity and magneto-resistivity as a function of temperature was measured down to 20 K by employing linear four probe technique.

Temperature variations of linear (χ_1' , χ_1'') and third harmonic ac susceptibility (χ_3' , χ_3'') were measured down to 20 K by using mutual inductance bridge method. The in-phase and out-of-phase susceptibility signals were measured simultaneously using a dual phase lock-in amplifier. Temperature variations of χ_1' and χ_1'' were measured at five different frequencies (333, 1333, 3333, 6333 and 9333 Hz) at an ac field amplitude of 0.6 mT.

3. Results and discussion

Typical XRD patterns recorded for $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$ compounds with $y = 0.10, 0.30$ and 0.50 are shown in Fig. 1. The XRD patterns of samples for $y \leq 0.20$ could be indexed to $R\bar{3}c$ space group and the patterns for $y \geq 0.30$ could be indexed to $Pbnm$ space group. The XRD patterns were analyzed with the help of Fullprof program by employing Rietveld refinement technique. The typical lattice parameters for $y = 0.30$ sample are found to be $a = 5.4803(15) \text{ \AA}$, $b = 5.5275(17) \text{ \AA}$ and $c = 7.7785(28) \text{ \AA}$ and they are comparable to those reported for $\text{La}_{0.67}\text{Ca}_{0.33}\text{Mn}_{1-x}\text{Co}_x\text{O}_3$ [10]. The average valencies of Mn and Co determined from chemical titration suggest that there is a possible mixture of $\text{Mn}^{3+}/\text{Mn}^{4+}/\text{Co}^{4+}/\text{Co}^{3+}/\text{Co}^{2+}$ ions. The sample compositions estimated from SEM-EDS analysis are found to be comparable to the nominal starting compositions.

The temperature variations of χ_1' , the in-phase linear ac susceptibility for $y = 0.10, 0.15, 0.30$ and 0.50 are shown in Fig. 2. They exhibit PM–FM transition upon cooling. The

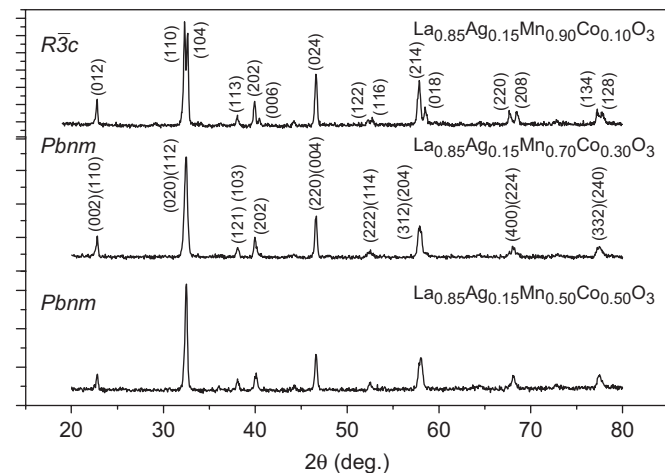


Fig. 1. XRD patterns of $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$ samples for $y = 0.10, 0.30$ and 0.50 . The patterns could be indexed to $R\bar{3}c$ for $y = 0.10$ and $Pbnm$ for $y = 0.30$ and 0.50 .

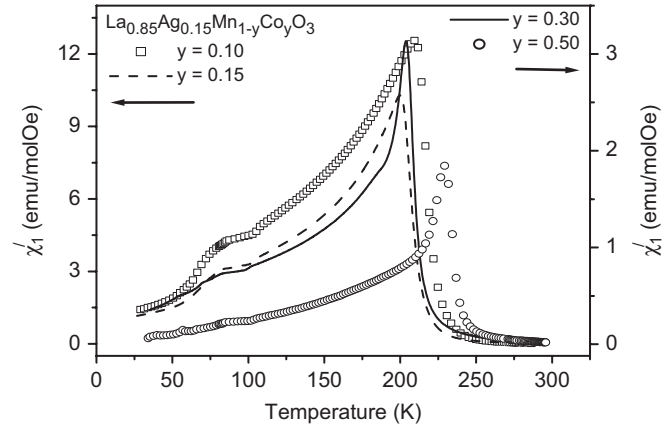


Fig. 2. Temperature variation of linear in-phase ac susceptibility (χ_1') of samples $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{1-y}\text{Co}_y\text{O}_3$ for $y = 0.10, 0.15, 0.30$ and 0.50 .

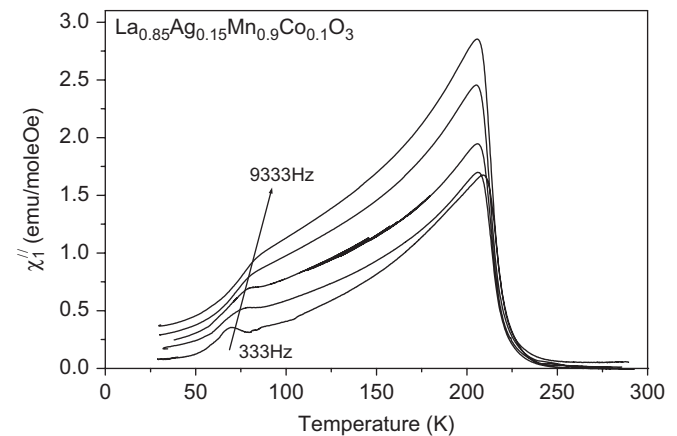


Fig. 3. χ_1'' versus temperature (T) measured at frequencies $f = 333, 1333, 3333, 6333$ and 9333 Hz for the sample $\text{La}_{0.85}\text{Ag}_{0.15}\text{Mn}_{0.90}\text{Co}_{0.10}\text{O}_3$.

FM transition could be understood due to the double exchange (DE) interaction between $\text{Mn}^{3+}-\text{O}^{2-}-\text{Mn}^{4+}$ networks. The sharp drop observed in χ_1' versus temperature plot below the FM transition could be due to possible FM–AFM transition. The FM T_c is found to decrease from 284 K for $y = 0$, to 199 K for $y = 0.20$ and for $y > 0.20$; it is found to increase systematically to a value of 234 K for $y = 0.50$. The initial decrease in T_c could be due to the dilution of $\text{Mn}^{3+}-\text{O}^{2-}-\text{Mn}^{4+}$ networks by the doped Co ions and the possible AFM interaction between Mn and doped Co ions. The increase in T_c for $y > 0.20$ could be due to DE FM interactions in $\text{Co}^{3+}-\text{O}^{2-}-\text{Co}^{4+}$ networks. Similar behavior was observed by Phuc et al. [17] in cobalites $\text{La}_{0.70}\text{Sr}_{0.30}\text{Co}_{1-y}\text{Mn}_y\text{O}_3$.

Typical plots of χ_1'' versus temperature for different frequencies are shown in Fig. 3 for $y = 0.10$ sample. We can see that other than the major peak in the vicinity of FM T_c , there is a minor peak at around 75 K. The minor peak shifts towards higher temperature with increase in frequency. These features are commonly observed in conventional spin-glass system. Similar frequency

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