

## Colossal magnetoresistance in layered manganite $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ ( $0 \leq x \leq 0.5$ )

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**Abstract.** The layered manganite  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ , with  $x$  varying between 0 and 0.5, has been synthesized using solid-state reaction method. We have found that  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  do not form the single-phase layered compound with  $\text{A}_3\text{B}_2\text{O}_7$  structure. Instead, mixtures of various phases, such as, orthorhombic perovskite, i.e.,  $\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$ , layered manganite and unreacted starting compounds, have been obtained. Except for  $x=0.4$ , which is found to be an antiferromagnetic insulator, all other  $x$  values yielded metal–insulator transition and ferromagnetic ordering.

**Keywords.** Colossal magnetoresistance; layered manganites;  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ .

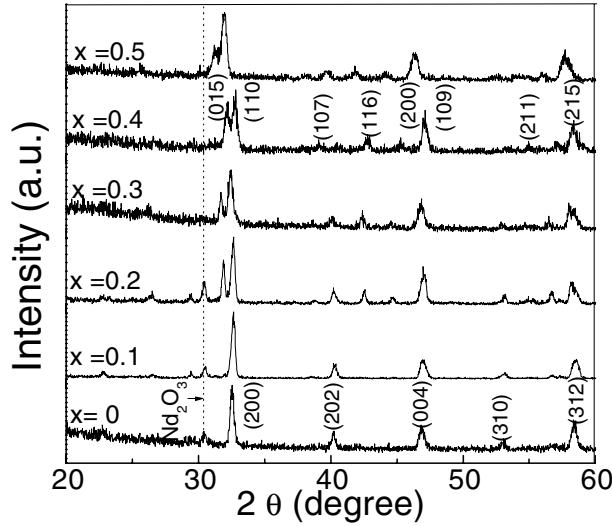
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### 1. Introduction

The observation of colossal magnetoresistance (CMR) in layered manganites, which is  $n = 2$  phase of Ruddlesden–Popper (RP) series, i.e.,  $\text{A}_3\text{B}_2\text{O}_7$  structure (space group  $I4/mmm$ ), has attracted considerable interest because of their two-dimensionality [1]. So far, majority of the research work has been centered on  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ . This material, for  $x = 0.3–0.45$ , exhibits an insulator–metal transition ( $T_{\text{IM}}$ ) at  $\sim 130$  K, and a ferromagnetic ordering at temperature  $T_c$ , which is much higher than  $T_{\text{IM}}$ . The observation of  $T_c \gg T_{\text{IM}}$  suggests that the double exchange mechanism alone does not explain the CMR in this material. On the other hand, the analogous compound  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  has not been studied in detail. Interestingly, this material, for  $x = 0.45$ , is found to be an antiferromagnetic insulator but exhibited a CMR over a wide range of temperature [2]. However for  $x = 0.3$ , the material showed IM transition [3] and ferromagnetic ordering. In this paper, we report on the synthesis of  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  series, with  $x$  varying between 0 and 0.5, and study their transport and magnetic properties.

### 2. Experimental

Polycrystalline  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  samples were prepared using standard solid-state reaction method. Stoichiometric quantities of  $\text{Nd}_2\text{O}_3$ ,  $\text{MnO}_2$  and  $\text{SrCO}_3$  were calcined at



**Figure 1.** XRD plots recorded for  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  samples with different  $x$  values. The dotted line shows the peak corresponding to impurity phase  $\text{Nd}_2\text{O}_3$ .

1200°C for 48 h. The material was reground, pelletized and sintered at 1250°C for 24 h. This process was repeated twice to obtain the homogeneous material. The phase identification in each case was carried out using X-ray powder diffraction (XRD). The magnetotransport measurements were carried out under an applied magnetic field of 1 T using standard four-probe resistivity technique.

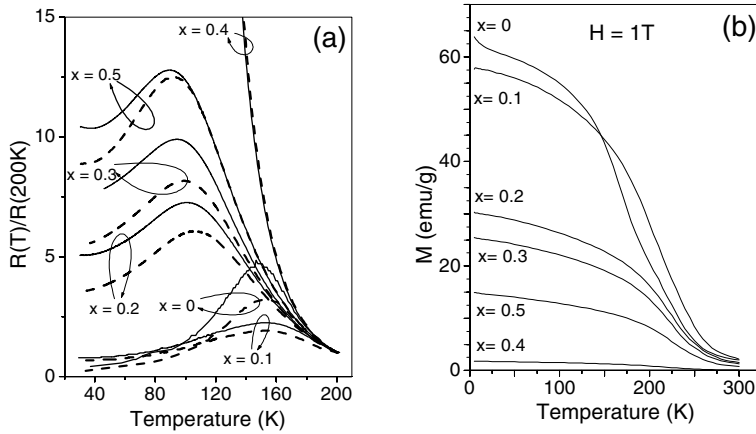
### 3. Results and discussion

The XRD patterns recorded for  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ , with different  $x$  values, are shown in figure 1. It is seen that single-phase material with  $\text{A}_3\text{B}_2\text{O}_7$  structure has not been formed for any of the  $x$  values. The lattice parameters for the major phase formed in each case were determined using the least square fitting and, the results are shown in table 1. For  $x = 0$  and 0.1, the major phase formed is orthorhombic perovskite  $\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$ . The layered  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  phase with orthorhombic structure, as a major phase, is formed for  $x$  values between 0.2 and 0.4. The orthorhombicity  $\{(a-b)/(a+b)\}$  is found to be minimum for  $x=0.4$ . However, for  $x=0.5$  again  $\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$  phase is seen to be the major phase.

The temperature dependence of normalized resistance in zero and 1 T magnetic field is shown in figure 2a. It is seen that except for  $x = 0.4$ , all the samples exhibit an insulator-to-metal transition and a negative magnetoresistance. The  $T_{\text{IM}}$  is found to decrease with  $x$ . For  $x = 0$  and 0.1, the highest MR was obtained in the vicinity of  $T_{\text{IM}}$ , while for other compositions a monotonous increase in MR with decreasing temperature was observed. The temperature dependence of magnetization for  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  samples is shown in figure 2b. Except for  $x = 0.4$ , which has an antiferromagnetic behavior, all other samples exhibit paramagnetic to ferromagnetic transition. The  $T_c$  was found to be  $\sim 275$  K, which

**Table 1.** Lattice parameters of the major phase formed during synthesis of  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ .

$x$	Major phase	$a$ (Å)	$b$ (Å)	$c$ (Å)
0	$\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$	5.434	5.801	7.698
0.1	$\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$	5.45	5.812	7.697
0.2	$\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$	3.844	3.932	20.137
0.3	$\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$	3.844	3.91	20.137
0.4	$\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$	3.847	3.904	20.029
0.5	$\text{Nd}_{1-z}\text{Sr}_z\text{MnO}_3$	5.466	5.697	7.829

**Figure 2.** Temperature dependence of (a) normalized resistance (solid lines in zero field and dotted lines in 1 T magnetic field) and (b) magnetization for  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  samples with different  $x$  values.

is much higher than  $T_{\text{IM}}$ . As mentioned in the introduction,  $T_c \gg T_{\text{IM}}$ , which has also been observed for  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ , and several explanations for this have been proposed and are a subject of debate at present. The proposed explanations basically fall into two categories. In the first category, the observation of  $T_c \gg T_{\text{IM}}$  has been attributed to the intrinsic character of this layered material. According to this explanation, the  $\text{MnO}_2$  layers act like 2D ferromagnets at temperatures between  $T_{\text{IM}}$  and  $T_c$  and 3D ordering takes place around  $T_{\text{IM}}$  [4]. The second category of explanations suggests that the higher  $T_c$  is due to extrinsic phases [5]. Our results indicate that the difference in  $T_c$  and  $T_{\text{IM}}$  arises from the second mechanism. However, more experiments are needed to resolve the issue.

#### 4. Conclusion

The layered manganite  $\text{Nd}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$  ( $x = 0.0-0.5$ ) has been synthesized and characterized for transport and magnetic properties. It has been found that this material does not form a single phase with layered  $\text{A}_3\text{B}_2\text{O}_7$  structure. For  $x = 0.4$ , the material is antifer-

romagnetic insulator and does not show CMR properties. For all other  $x$  values, the CMR properties arise due to the presence of extrinsic phases.

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