

MAGNETOTRANSPORT STUDIES OF $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ PREPARED BY SOL GEL METHOD

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ABSTRACT

The manganite oxide, $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ was prepared by sol-gel method in order to obtain high purity homogeneous powders and to lower the temperature values of heat treatments. The material is characterized by powder X-ray diffraction, temperature dependence of ac magnetic susceptibility, resistivity and magnetoresistance measurements in a magnetic field of 1 Tesla. X-ray powder diffraction showed that the sample is single-phased compound. AC susceptibility and resistivity measurements show that the ferromagnetic phase transition at 200 K with high magnetoresistance of 20 % at transition temperature (T_P). The parent compounds $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ ($\langle r_A \rangle = 1.2365$, $T_N = 150$ K) and $\text{Nd}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ ($\langle r_A \rangle = 1.1715$, $T_N = 160$ K) are charge ordered antiferromagnetic insulators. However, $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ ($\langle r_A \rangle = 1.1975$) is a ferromagnetic metal. This dramatic change is attributed to an increase of A site cation disorder, which changes the e_g -electron transfer interactions.

KEYWORDS: Colossal magnetoresistance, manganites, charge ordering, sol-gel method

1. INTRODUCTION

The colossal magnetoresistance materials, $\text{A}_{1-x}\text{B}_x\text{MnO}_3$ (A-rare earth; B-alkaline earth) have attracted much interest due to their potential technological applications and the intriguing physics involved. Different substitutions in the manganites lead to different crystal structures, magnetic and electronic phase transitions resulting in a complex phase diagram. These phenomena can be explained by a combination of double exchange mechanism¹, electron-phonon coupling², super-exchange, orbital ordering, charge ordering³ and Jahn–Teller interactions⁴. The average radius of the A site cation ($\langle r_A \rangle$) strongly influences the magnetic and transport properties of manganites.

In the present study carried out, the influences of $\langle r_A \rangle$ on electric and magnetic properties of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ compound with the help of resistivity, ac susceptibility and magnetoresistance measurements were explored. The parent compound $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ transforms a paramagnetic metallic state to ferromagnetic metallic at a temperature $T_C = 250$ K, and at a temperature of about 158 K to an antiferromagnetic insulating charge-ordered state⁵. In $\text{Nd}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ antiferromagnetic charge ordering occurs around 160 K⁶. It has been shown that the change in $\langle r_A \rangle$ drives antiferromagnetic parent compounds to ferromagnetic compound.

2. EXPERIMENT

$\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ was prepared by citrate gel (sol gel)⁷ method. This method is known to give a high degree of homogeneity since the produced oxides are acquired from a solution of their precursors. The starting materials Nd_2O_3 , SrCO_3 and CaCO_3 were first dissolved in dilute nitric acid, $\text{Mn}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$ in water and then appropriate mixture of citric acid. The

resulting gel was decomposed at 300 °C and the acquired precursor powder was heated in air at 600 °C for 6 hours and then at 1100 °C for 24 hours to improve crystalline nature.

Powder XRD patterns of the sample were recorded by using Shimadzu-D X-ray diffractometer, with CuK_α radiation. The spectra were obtained in the angle range $2\theta = 5^\circ\text{--}100^\circ$ with step size of 0.02. Resistivity measurement was performed using the conventional four probe method in the temperature range 35-300 K. A Janis closed cycle refrigerator was used for cooling the sample. Magnetoresistance of the sample was measured by four-probe method using a liquid nitrogen gas flow cryostat (77-300 K) with magnetic field of 1 Tesla. AC susceptibility measurement below room temperature was performed using a Sumitomo ac susceptometer with magnetic field of 10^{-3} Tesla and at a frequency of 313 Hz, in the temperature range 18-300 K.

3. RESULTS AND DISCUSSION

The X-ray diffractograms of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ is given in figure 1. Pattern is analyzed by Rietveld refinement technique, which confirmed that the sample is single phase and orthorhombic with the space group Pnma. Refinements were done on the background using a Shifted Chebyshev polynomial. The oxygen positions and thermal parameters were not refined. Lattice parameters, goodness of fit and calculated $\langle r_A \rangle$ of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ are given in the Table I. For the comparison, the corresponding values for $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ and $\text{Nd}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ are also given. The values of lattice parameters a and c of the sample lie between those of the parent compounds. But the value of b is larger than in the parent compounds, which indicates that distortion of lattice causes an elongation of b . This might indicate the preferential occupation of e_g electrons in the d_{z^2} orbitals which increases the e_g transfer interaction and favors the ferromagnetic interaction along the b directions. The unusual behavior of phase transition occurs for an intermediate range of $\langle r_A \rangle$ from 1.1715 Å to 1.2365 Å. Inhomogeneous local distortion of lattice in this range of $\langle r_A \rangle$ leads to a phase transition from antiferromagnetic parent compounds to ferromagnetic compound.

Resistivity (ρ) versus temperature plot for the sample is given in figure 2. The resistivity shows a metallic behavior below transition temperature ($T_p=175$ K) and a semi conducting-like behavior above transition temperature. The temperature corresponding to the resistivity peak T_p is 175 K. The temperature variation of the ac susceptibility, χ , of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ are shown in figure 3. It is seen that the susceptibility exhibits a sharp increase below 200 K indicating an occurrence of ferromagnetic ordering, which is consistent with the previous report⁸. Ac susceptibility values show there is no antiferromagnetic transition compared to parent compounds. Hence, variation $\langle r_A \rangle$ drastically changes the magnetic ordering. The magnetoresistance of the sample in 1 Tesla field is shown in the figure 4. There is a peak at the transition temperature with negative magnetoresistance ratio of 20%. In the temperature range 212-272 K, it shows a constant MR of 5%. MR is found to have large values in low temperature region, which is attributed to tunneling across the grain boundaries⁹.

4. CONCLUSION

In summary, magnetic and transport properties of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ completely modified from the parent compounds which is attributed to change in e_g transfer interaction due to A site cation disorder. In this paper, we report the preliminary results of this compound showing reentrant ferromagnetism.

References

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TABLES

Table I

Parameters	Nd _{0.5} Ca _{0.5} MnO ₃	Nd _{0.5} Ca _{0.3} Sr _{0.2} MnO ₃	Nd _{0.5} Sr _{0.5} MnO ₃
R _{WP}	4.90%	5.40%	7.23%
R _p	3.35%	4.5%	4.65%
2 θ Range	10-60°	10-100°	10-82°
a (Å)	5.39200(6)	5.41287(5)	5.43153(3)
b (Å)	7.58972(8)	7.66720(3)	7.63347(4)
c (Å)	5.37605(6)	5.40216(0)	5.47596(3)
space group	Pnma	Pnma	Imma
volume(Å ³)	220.008(5)	224.198(0)	227.041(2)
<r _A > (Å)	1.1715	1.1975	1.2365

FIGURES

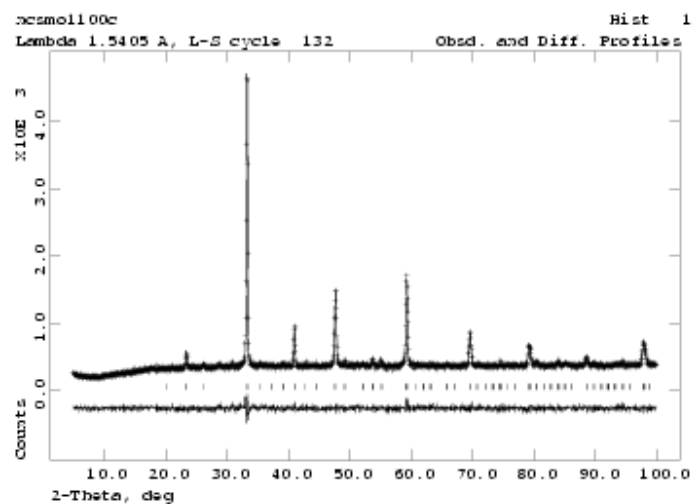


FIG.1: Rietveld refinement profile for $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_{0.2}\text{MnO}_3$ sample performed using GSAS.

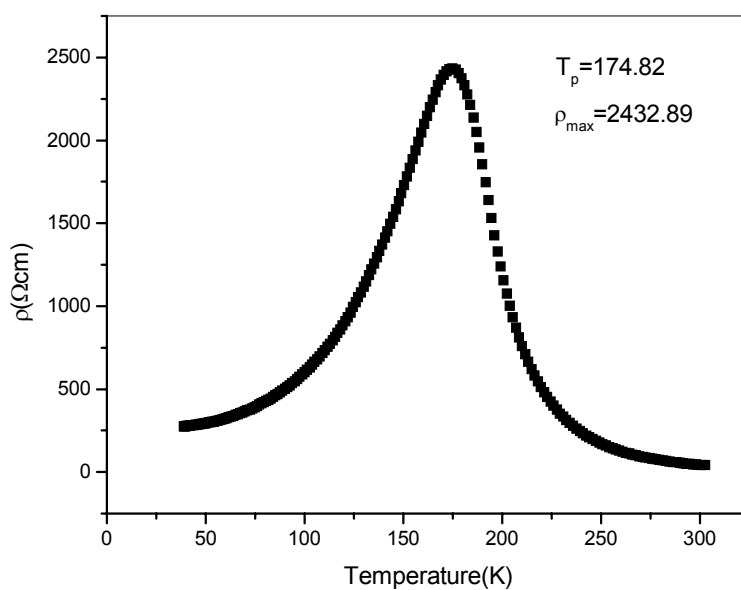


FIG. 2: Resistivity of the sample as a function of temperature.

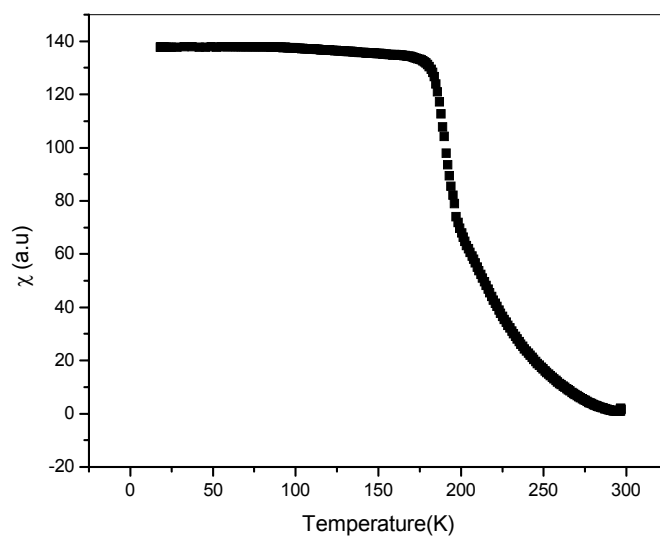


FIG. 3: AC susceptibility of $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_2\text{MnO}_3$ with magnetic field of 10^{-3} Tesla.

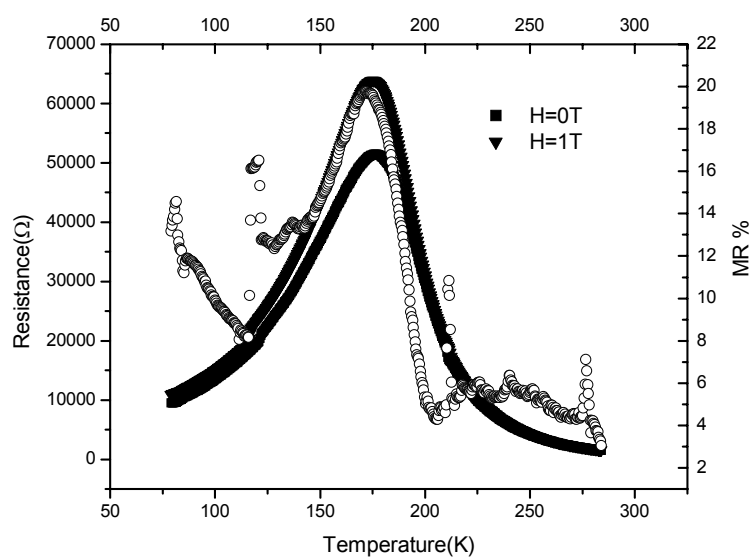


Fig. 4: Resistance vs. Temperature in $H = 0$ and 1 Tesla and magnetoresistance for $\text{Nd}_{0.5}\text{Ca}_{0.3}\text{Sr}_2\text{MnO}_3$.