



INFLUENCE OF A-SITE DISORDER ON THE MAGNETORESISTANCE IN $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ AND $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ MANGANITES

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ABSTRACT

Analysis of magnetoresistance above and below Curie temperature (T_C) in $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ (PSMO) and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ (PHSMO) manganites is reported. T_C of the PSMO sample was found to be higher than the T_C of PHSMO. The magnetoresistance above T_C was found to scale as the square of the Brillouin function and below T_C as the Brillouin function. K , the average moment per hopping site was found to be higher for the PHSMO sample compared to the PSMO sample. These results are attributed to the increase in A-site disorder for PHSMO sample, which increases the tendency to form clusters and decreases T_C .

Keywords: Manganites, Magnetoresistance, Brillouin function

1. INTRODUCTION

The perovskite type Colossal Magnetoresistance (CMR) manganites of general composition $\text{R}_{1-x}\text{A}_x\text{MnO}_3$, where R is a rare earth ion and A is a divalent alkaline earth ion, exhibit an unusually strong interplay between orbital, lattice and spin degrees of freedom. At temperatures below their Curie temperature (T_C), optimally substituted ($x \sim 0.3$) manganites are ferromagnetic metals, while at higher temperatures, i.e. above T_C , they exhibit a paramagnetic insulating behaviour¹. This generic behaviour, as well as the magnetoresistive effect which occurs near the metal-insulator transition (MIT), has been understood within the framework of the Zener double exchange (ZDE) model^{2,3}. The transfer (hopping) of an e_g electron between the neighbouring Mn^{3+} and Mn^{4+} ions through the Mn-O-Mn path, results in an effective ferromagnetic interaction due to strong on-site Hund's coupling. When manganites are doped with divalent alkaline earth ions like Ca^{2+} , Sr^{2+} etc. at the A-site, the carrier concentration as well as the average A-site ionic radius changes. On the other hand, when isovalent rare earth ion like Ho^{3+} is substituted at the A-site, only the average A-site ionic radius changes. The change in the A-site ionic radii affects the ferromagnetic exchange mechanism, which causes a change in the ferromagnetic transition temperature (T_C) as well as the resistivity and magnetoresistance (MR)⁴⁻⁹. The average A-site disorder is characterized by a parameter $\langle \sigma \rangle^2 = \sum_i y_i r_i^2 - \langle r_A \rangle^2$

where r_i is the ionic radius of the A site ion of concentration y_i . In this work, we report on the effect of this A-site disorder on the magnetoresistance of $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ (PSMO26) and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ (PHSMO). A very small concentration of Ho ($=0.04$) was chosen to study the subtle variations in transport properties brought about by this substitution.

2. EXPERIMENTAL DETAILS

Polycrystalline samples of the nominal composition $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ (PSMO26) and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ (PHSMO) were prepared using the conventional solid-state method. X-ray diffraction (XRD) of the samples was performed using $\text{Cu-K}\alpha$ radiation (Rich-Seifert, Germany). The electrical resistivity of the samples as a function of both temperature (2K to 400K) and field (-7T to 7T) was measured using the standard linear four-probe technique in the temperature range of using an Oxford continuous flow cryostat. Magnetization measurements were carried out using a SQUID magnetometer (Quantum design MPMS XL) in the same temperature range in a field-cooled mode of 1000 Oe on powdered samples.

3. RESULTS AND DISCUSSIONS

X-ray diffraction pattern of the samples are shown in fig.1. Both the samples could be indexed to a pseudo-cubic structure. The Curie temperature T_C which is temperature in which the temperature derivative of magnetisation is minimum, is seen to be higher for the PSMO26 sample (204K) than the phsmo sample (159K) (figure 2). It is also seen that the peak resistivity temperature T_P for the phsmo sample is higher than that of the PSMO26 sample and so is the value of the resistivity (figure 3). This can be explained as follows. Ho^{3+} ion has a smaller ionic radii (1.072Å) compared to the Pr^{3+} (1.179Å) and Sr^{2+} (1.31Å)¹⁰. Therefore, substituting Ho^{3+} ions in the place of Pr^{3+} ions decreases $\langle r_A \rangle$ the average ionic radii, and increases $\langle \sigma^2 \rangle$ ($\langle \sigma^2 \rangle = 3.66 \times 10^{-3} \text{Å}^2$ for the PSMO26 sample and $4.34 \times 10^{-3} \text{Å}^2$ for the PHSMO sample). This leads to a decrease in the Mn-O-Mn bond angle away from 180° leading to a lowering of the hopping integral and hence narrowing of the bandwidth. These changes affect the transfer of the e_g electrons and therefore results in a lower T_C and T_P and increase in resistivity value. A similar behaviour is seen in other rare earth ion doped manganite samples also where a smaller rare earth replaces Pr^{3+} ion in the A-site¹¹. The magnetoresistance of the samples are plotted in fig.4a and 4b. It was analysed using a model proposed by Wagner *et al*¹². They have analysed their magnetoresistance using a modified form of Mott's variable range hopping (VRH) mechanism. In this model the hopping barrier W_{ij} is modified by an extra ΔW_{ij} which is the energy related to the relative orientation of the magnetization vectors \vec{M}_i and \vec{M}_j at both sides of the hopping process. This hopping barrier is enhanced for a paramagnet and is reduced for a ferromagnet.

The magnetoresistance can be written therefore as $\frac{\Delta\rho(B,T)}{\rho_0} \approx \frac{\Delta W_{ij}}{k_B T}$, where ΔW_{ij} is proportional to

$B_K^2 \left(\frac{gK(T)\mu_B H}{k_B T} \right)$ (where $K(T)$ is the average moment between the hopping sites), the square of the

Brillouin function in the paramagnetic state and to constant $A(T)$ is related to $\rho_0(T)$ which is the resistivity in zero magnetic field.

The fits to these is given in fig. 4a and 4b and the corresponding temperature variations of the parameters $A(T)$ and $K(T)$ are given in fig 5. From fig 5. it is clear that the value of K the average moment between the hopping sites increases as we approach T_C and gradually goes to zero as the sample is cooled below T_C towards lower temperature. This behaviour is consistent with the model of Wagner and co-workers because as the sample is cooled down to lower temperatures the Weiss field contribution \vec{M}_w becomes comparable to the saturation magnetization \vec{M}_s and hence the local correction to the magnetization $\delta M_{i,j}$ goes to zero. In this scenario, the carriers are mobile and delocalized within each individual cluster while the macroscopic resistivity is determined by the magnetically impeded transfer of carriers between

adjacent spin clusters. It is interesting to note that the value of K , the average spin moment per hopping site is higher for the phsmo sample compared to the psmo26 sample. As seen earlier the PHSMO sample has a higher $\langle\sigma^2\rangle$ compared to the psmo26 sample i.e. the undoped sample.

This increase in the variance in A-site disorder leads to an additional trapping of carriers in the lattice due to the enhanced disorder brought about by the Ho^{3+} doping at the A-site. This leads to an inhomogeneity in the lattice with carriers having different mobilities and results in the formation of magnetic spin clusters. These clusters extend upto a few lattice constants and coalesce into an infinite cluster near T_C ¹³. Thus it is seen that even substituting a very small amount of Ho^{3+} ion in place of Pr^{3+} ion brings about a tangible change in the physical properties in these materials showing that disorder plays a main role in controlling the physics in these systems.

4. CONCLUSIONS

We have studied the temperature and field variation of resistivity both above and below the Curie temperature T_C in $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ (PSMO26) and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ (PHSMO) manganite samples. It is seen that both the peak resistivity temperature and the Curie temperature is higher for the Ho doped sample compared to the undoped psmo26 even for a very small concentration of 0.04 of Ho. This is attributed to the increase in the variance of the A-site disorder due to the difference between the ionic radii of Ho^{3+} and Pr^{3+} ions. The magnetoresistance above T_C was found to scale as the square of the Brillouin function while below T_C as the Brillouin function. The average moment per hopping site was found to peak near T_C with its value being higher for the phsmo sample than the PSMO26 sample. This is due to the enhancement in the localization of carriers due to the increase in variance of the disorder in A-site which results in the formation of spin clusters.

5. REFERENCES

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FIGURES

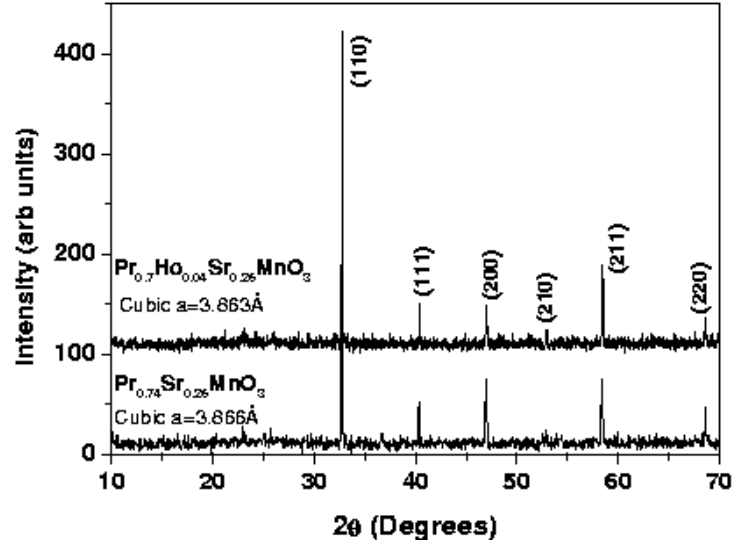


Figure 1: The X-ray diffractograms of $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$. The samples could be indexed to a pseudo cubic structure.

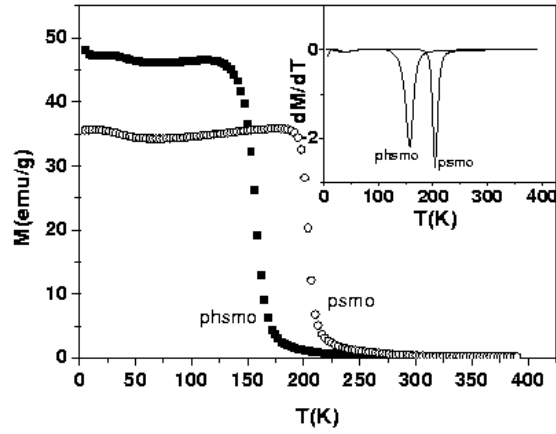


Figure 2: The Temperature variation of magnetisation for the two samples. Inset shows the temperature derivative of magnetisation whose minimum is taken to be the Curie temperature T_C .

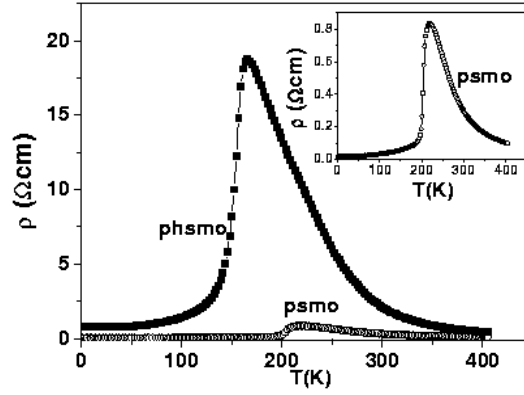


Figure 3: Temperature variation of resistivity for both the samples. The Ho substituted sample has about an order high magnitude of resistivity owing to higher A-site disorder. The inset shows the resistivity of the $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ sample alone for clarity sake.

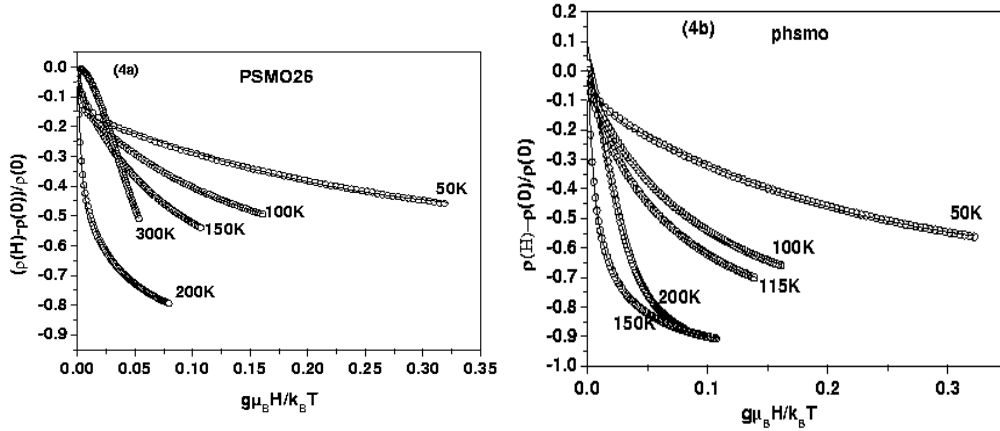


Figure 4: 4a. shows the Brillouin fits of magnetoresistance for $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ sample while the corresponding fits for the $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ is shown in Fig. 4b. Symbols represents data points while the line are the fits to the Brillouin function (below T_C) and to the square of the Brillouin function above T_C .

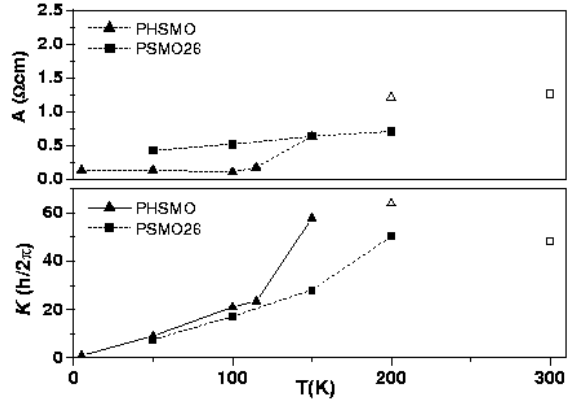


Figure 5: represent the temperature evolution of the fit parameters A and K for $\text{Pr}_{0.74}\text{Sr}_{0.26}\text{MnO}_3$ and $\text{Pr}_{0.7}\text{Ho}_{0.04}\text{Sr}_{0.26}\text{MnO}_3$ samples. The open symbol represents the point in the paramagnetic regime (above T_C) while the closed symbol represents the ferromagnetic regime (below T_C).