THEORETICAL STUDY OF MAGNETIC SUSCEPTIBILITY OF COLOSSAL MAGNETO RESISTIVE MANGANITES ($Re_{1-x}\,A_x\,MnO_3$); A VARIATIONAL TREATMENT

Amit Chaudhary[1], Sunil Panwar[2], Rajendra Kumar[3]

[1,3]Department of Physics, Gurukula Kangri University, Haridwar, Uttarakhand, (India)

[2] Department of Applied Physics, Faculty of Engineering & Technology, Gurukula Kangri University, Haridwar, Uttarakhand, (India)

ABSTRACT

In this work, we use a simple variational method for calculating the magnetic susceptibility(X_S) of rare earth manganites doped with alkaline earths namely $Re_{1-x}\,A_x\,MnO_3$ (where Re = La, Pr, Nd etc. and A= Ca, Sr, Ba etc.) which exhibit colossal magnetoresistance (CMR), metal insulator transition & many other poorly understood phenomena. We have recently studied a two band model Hamiltonian for manganites in the strong electron- lattice Jahn-Teller (JT) coupling regime. Using this Hamiltonion, we have studied the role of the model parameters e.g. local coulomb repulsion U, Hund's coupling J_H between e_g spins & t_{2g} spins, ferromagnetic nearest neighbor exchange coupling J_F between t_{2g} core spins & hybridization V_k between ℓ -polarons & d-electrons in the behavior of the magnetic susceptibility X_S of these materials. We find that the maximum of X_S occurs at a temperature T^* (~ 200 K) followed by a sudden drop in X_S as we decrease the temperature. This behavior resembles with the key feature of many CMR compounds like $La_{1-x}\,Ba_x\,MnO_3$ with x=0.02-0.35 & $La_{0.67}\,Ca_{0.33}\,Mn_{0.9}\,Ta_{0.1}O_3$. This anomaly in X_S arises from the onset of magnetic ordering at 200K and decreases on increasing V_k or J_H value. While it increases with increasing J_F value and the peak at low temperature becomes more sharpened.

Keywords: Colossal Magneto resistive Manganites, Magnetic Susceptibility, Magnetic Properties, Variational Techniques

I. INTRODUCTION

Doped perovskite manganites $Re_{1-x}\,A_x\,MnO_3$ (where Re and A are rare earth & alkaline earth ions) have been extensively studied over the last decade. This system is found to exhibit fascinating properties [1, 2] such as metal-insulator transition (MIT), ferromagnetic (FM)- paramagnetic (PM) phase transition, colossal magnetoresistance (CMR), charge and orbital ordering etc. Previously, the transport and magnetic properties of the manganites were explained by the double-exchange (DE) mechanism [3] combining with the local Jahn-Teller distortions of Mn^{3+} ions [4].However, due to various interactions among charge, spin and lattice these materials become more complex and DE alone cannot explain the entire electrical transport behavior. Later on various theoretical models have been proposed by considering electron-lattice & spin lattice interaction & even today there is no comprehensive model to explain transport phenomena in manganites [4, 5]. Recently, it has been found that manganese oxide display a rich phase diagram [6, 7]. Percolation based on phase separation has been proposed to explain magnetotransport properties in these systems [8]. There are many excellent review articles [9-13] & edited books [14, 15] which discuss the evidence of coexistence of different electronic & magnetic (FM & AFM or PM) as well as charge and/ or orbitally (metallic & insulating) ordered phases in manganites. A phenomenon of phase separation has been discussed in manganites by a review by Dagotto, Hotta & Mereo [16]. In it, they explained theoretically the possibility of electronic phase separation between anti ferromagnetic insulating & ferromagnetic metallic states. Recently Ramakrishnan et al.[17] has carried the detailed study of rare earth manganites doped with alkaline earths namely $Re_{1-x} A_x MnO_3$ which exhibit CMR , metal insulator transition & many other poorly understood phenomena where the authors presented a new model of coexisting localised JT polarons and broad band electrons for manganites and argued that it arises inevitably in the presence of orbital degeneracy and strong JT coupling and shown that it explains a wide variety of characteristic properties of manganites. The theory ignores the ℓ-b hybridization so it does not describe the low temperature behavior of manganites below T*~ 100 K. We believe that a more general treatment of the model which includes ℓ-b hybridization can lead to a complete description of manganites as suggested by Ramakrishanan et al. (Ref. 17).

We developed some time ago a variational method to study the ground state & thermo-dynamic properties of heavy fermion systems using periodic Anderson model [18]. The temperature dependence of the magnetic susceptibility X_S (T) of doped CMR manganites have been studied in the present paper using this variational technique.. We had already used this variational method in the study of the zero field electrical resistivity of doped CMR manganites over a fairly wide temperature range [19]. In Section 2, we give the basic formulation for X_S (T). In Section 3, we discuss our results.

II. BASIC FORMULATION

2.1 Model Hamiltonion

We take here the two band model Hamiltonian suitable for the doped manganites which exhibit colossal magnetoresistance (CMR) involving a broad spin- majority ($\boldsymbol{e_g}$-spins) conduction band **(b-band)** as well as nearly localized spin- minority($\boldsymbol{t_{2g}}$-spins) electron states(**ℓ-band**).Two band models involving itinerant & localized states were also suggested earlier by both experimentalists & theorists [20-22]. In the two- band picture, we start with a model Hamiltonian which includes l-b hybridization effects and thus can address the low temperature properties of manganites (e.g. resistivity, Hall effect) [17, 19] given by-

$$\begin{aligned} H_{lb} = & -\sum_{\langle ij\rangle\sigma} \bar{t}_{ij}\left(b^{+}{}_{i\sigma}\, b_{j\sigma}\right) - \sum_{i\sigma} E_{jt}\, l^{+}{}_{i\sigma}\, l_{i\sigma} + \mathrm{U}\sum_{i\sigma} n^{l}{}_{i\sigma}\, n^{b}{}_{i\sigma} - J_H \sum_{i} \boldsymbol{s}_i \cdot \boldsymbol{S}_i - J_F \sum_{\langle ij\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \\ & - \sum_{k\sigma} V_K \left(l^{+}{}_{k\sigma}\, b_{k\sigma} + h.c.\right) \\ & - \mu\mathrm{B} \sum_{\langle ij\rangle} \boldsymbol{S}_i . \mathbf{H} \qquad (1) \end{aligned}$$

The details of symbols used are given in our previous work in Ref. [19].In Eq. (1), the ℓ- polaron has energy ($-E_{JT}$), the b- electrons hope between nearest neighbor sites with an effective amplitude $\ddot{t}$. U is the local Coulomb repulsion between l- polarons & b- electrons of the same spin at a particular site i. V_k is the ℓ-b hybridization between l- polarons & b-electrons of the same spin. J_H is the strong ferromagnetic Hund's rule coupling between the e_g spin $\overrightarrow{s_i}$ & the t_{2g} spin $\overline{S_i}$. J_F is the net effective ferromagnetic nearest neighbor exchange coupling between the t_{2g} core spins (S_i, S_j) and the last term denotes the interaction of the latter with an external magnetic field **H** .

It has been shown in our earlier calculations on doped CMR manganites in Ref. [19] that in k- space, H_{lb} within mean field approximation may be written as

$$H_{lb} = -(E_{JT} + J_F\sigma m - \frac{\bar{U}}{2}n^b{}_\sigma - \frac{J_H}{2}n^b{}_{-\sigma})\sum_{k\sigma} l^+{}_{k\sigma}\, l_{k\sigma} + (\epsilon_k + \frac{\bar{U}}{2}n^l{}_\sigma)\sum_{k\sigma}(b^+{}_{k\sigma}\, b_{k\sigma})$$
$$+\frac{J_H}{2}n^l{}_\sigma\sum_{k\sigma}(b^+{}_{k-\sigma}\, b_{k-\sigma}) - \sum_{k\sigma} V_K(l^+{}_{k\sigma}\, b_{k\sigma} + h.c) \qquad (2)$$

Here

$$\epsilon_K = \sum_{ij} \tilde{t}_{ij}\, e^{ik}(R_i - R_j)\ ;\ R_i\ \&\ R_j\ \text{are the position vectors of } i\ \&j \text{ sites}\quad \&\ \bar{U} = U - J_H$$

and ferromagnetic exchange 'J_F' term of Eq.(1) within mean field approximation, reduces to

$$-J_F\,\sigma\, m\, \sum_{k\sigma} l^+{}_{k\sigma}\, l_{k\sigma}$$

where

$$m = \frac{1}{N}\sum_{k\sigma}\sigma l^+{}_{k\sigma}\, l_{k\sigma} \qquad (3)$$

is the magnetization per site & J_F involves the number of nearest neighbors [18].

2.2 Wave - Function

In the finite interaction U case, the modified variational wave-function [18] may be written as in k-space

$$\psi_{lb} = \prod_{k\sigma}\ [1 + A_{k\sigma}\, l^+{}_{k\sigma}\, b_{k\sigma}]\, |\Phi_d\rangle \qquad (4)$$

where $|\Phi_d\rangle$ is the Fermi sea of broad d-states & $A_{k\sigma}$ is the variational parameter.

When the energy expection value E is optimized with respect to the $A_{k\sigma}$'s, one finds

$$A_{k\sigma} = \frac{1}{2V_k}\left[(\epsilon_k + \frac{U}{2}n^l{}_\sigma - \epsilon_{d_\sigma}) + \sqrt{(\epsilon_k + \frac{U}{2}n^l{}_\sigma - \epsilon_{d_\sigma})^2 + 4V_k{}^2}\right] \qquad (5)$$

Where $\epsilon_{d_\sigma} = \frac{U}{2}\, n^b{}_\sigma + \frac{J_H}{2}\, n^l{}_\sigma - E_{JT} - J_F\sigma m \qquad (6)$

The number of b-electrons & l -electrons are given by

$$n^b{}_\sigma = \frac{1}{N}\sum_k n^b{}_{k\sigma} = \frac{1}{N}\sum_k \frac{f_{k\sigma}^-}{(1 + A_{k\sigma}{}^2)} \qquad (7)$$

$$\&\ n^l{}_\sigma = \frac{1}{N}\sum_k n^l{}_{k\sigma} = \frac{1}{N}\sum_k \frac{A_{k\sigma}{}^2 f_{k\sigma}^-}{(1 + A_{k\sigma}{}^2)} \qquad (8)$$

Here $f^-{}_{k\sigma}$ *is* the Fermi function for the lower branch of the quasi particle spectra $E_{k\sigma}^-$ given by

$$f_{k\sigma}^- = 1/\exp[\,\beta(E_{k\sigma}^- - \mu) + 1] \qquad (9)$$

Where μ is the chemical potential and $\beta = 1/k_B T$.

The expression for the lower branch of the quasi particle spectra is given by

$$E_{k\sigma}{}^- = \frac{1}{2}\left[\left((\epsilon_k + \frac{U}{2}n^l{}_\sigma + \epsilon_{d_\sigma}\right) - \sqrt{\left((\epsilon_k + \frac{U}{2}n^l{}_\sigma - \epsilon_{d_\sigma}\right)^2 + 4V_k{}^2}\right] \qquad (10)$$

2.3. Magnetic Susceptibility

The static magnetic susceptibility is given by

$$X_S = g\,\sigma\,\mu_B \frac{\partial}{\partial B}[n^{\ell}_{\sigma} - n^{\ell}_{-\sigma}\,]_{B \to 0} \qquad (11)$$

Putting n^{ℓ}_{σ}'s from eq. (8), one gets the expression of susceptibility X s in the units of $(g\,\mu_B)^2$ as

$$\chi_S\,(U, J_F) = \sum_{k\sigma}\left(\frac{\chi_0(U)}{[1+(U/2)I_3-((\frac{U}{2}+2J_F)\,\chi_0(U)/2]} \right) \qquad (12)$$

Where $$\chi_0(U) = I_1 + I_2 \qquad (13)$$

$$I_1 = \frac{2A_{k\sigma}\,V_K\,I_3}{\left[\sqrt{\left((\epsilon_k+\frac{U}{2}n^{l}_{\sigma}-\epsilon_{d_\sigma}\right)^2+4V_k^{\,2}}\right]} \qquad (14)$$

$$I_2 = \frac{4\,A^2_{k\sigma}/(1+A^2_{k\sigma})^2}{\left[\sqrt{\left((\epsilon_k+\frac{U}{2}n^{l}_{\sigma}-\epsilon_{d_\sigma}\right)^2+4V_k^{\,2}}\right]} \qquad (15)$$

$$I_3 = \frac{A^2_{k\sigma}}{(1+A^2_{k\sigma})}\quad \beta \exp[\beta\,(E_{\bar{k\sigma}} - \mu)]\,(f_{\bar{k\sigma}})^2 \qquad (16)$$

III. FIGURES

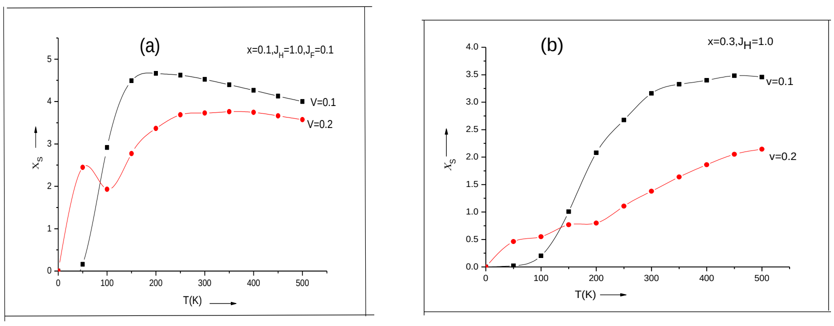

Fig.1 (a & b): Variation of magnetic susceptibility (X_S) with temperature T (K) at U=5.0,E_{jt}=0.5,J_H=1.0 & J_F=0.1 for different values of V with a) x=0.1, b) x=0.3

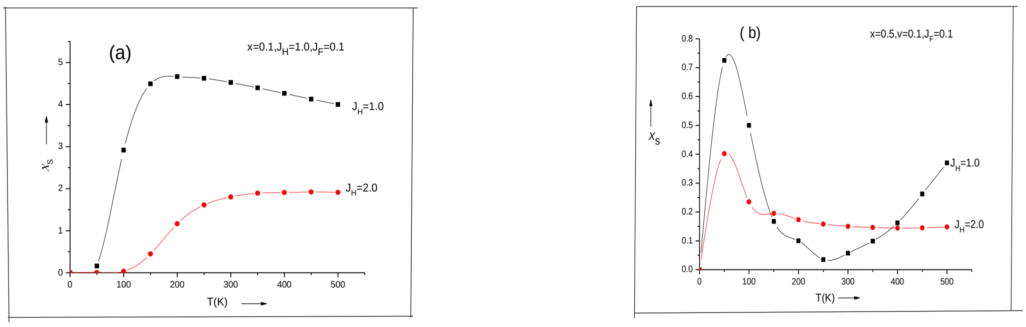

Fig. 2 (a & b): Variation of magnetic susceptibility (X_S) with temperature T (K) at U=5.0,E_{jt}=0.5 V=0.1 & J_F=0.1 for different values of J_H with a) x=0.1 & b) x=0.5

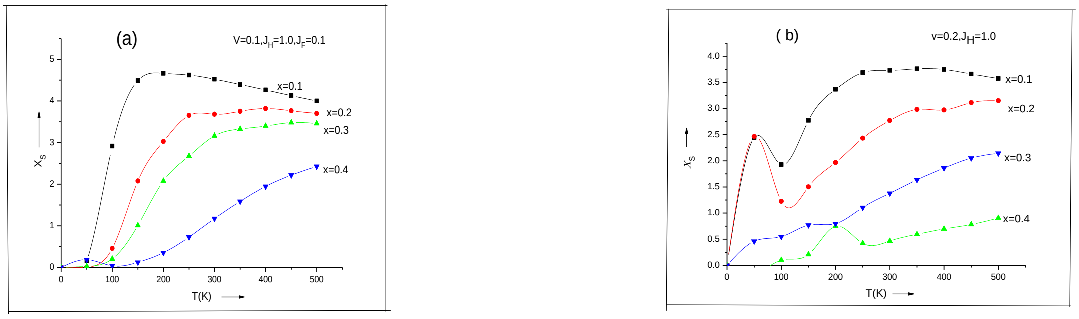

Fig .3(a & b): Variation of magnetic susceptibility (X_S) with temperature T (K) at U=5.0,E_{jt}=0.5 J_H=1.0 & J_F=0.1 for different values of x with a). v=0.1 & b). v=0.2

Fig. 4 (a & b): Variation of magnetic susceptibility (X_S) with temperature T (K) at U=5.0,E_{jt}=0.5 V=0.2 for different values of J_F with a). x=0.2 & J_H=2.0 & b). x=0.3& J_H=1.0

IV. RESULTS & DISCUSSION

In these calculations, we have considered the unperturbed band of three dimensional solid represented by simple semicircular density of states $N^c_\sigma(\epsilon_k) = (2/\pi)\sqrt{(1-\epsilon_k^2)}$ (which is centered around zero energy) with band width W=2.0eV, U=5.0 ,E_{jt}=0.5,V= 0.1& 0.2 , ϵ_F = -0.238 eV (for x =0.3) and J_H = 1.0 & 2.0eV. Doping concentration x is varied from 0.1 to 0.4.

In Figs. 1-4, we have shown the temperature dependence of magnetic susceptibility (X_S) for different values of parameters V, J_H, x & J_F . Here X_S follows a Curie- Weiss behavior at high temperatures well above T^* where the short-range FM fluctuations are negligible. The Curie- Weiss law is not followed towards lower temperature. At low temperature ,it shows a peak at T^* ~ 200 K (see fig.1 a) followed by a sudden drop in X_S as we decrease further the temperature resembling with the key feature of many CMR compounds like $La_{1-x}Ba_xMnO_3$ with x=0.02-0.35 [23]& $La_{0.67}Ca_{0.33}Mn_{0.9}Ta_{0.1}O_3$ [24].This sharp cusp like anomaly in X_S arises from the onset of magnetic ordering at 200 K & decreases on increasing V, J_H or doping concentration x . The ferromagnetic exchange interaction J_F enhances the X_S appreciably in the low temperature. We have seen that the results of the simple model considered here are in qualitative agreement with the experimental results of a broad class of hole doped CMR manganites.

V. CONCLUSION

The discovery of CMR effect in the mixed- valence hole doped manganites have attracted much attention for their scientific and technological interests. Magnetic perovskites (manganites) such as $La_{0.7}Sr_{0.3}MnO_3$ have attracted as potential magneto resistive sensors. CMR materials with integrated electronics have attracted as high sensitivity magnetic field sensors. While the applications of these materials are well known as magneto resistive (MR) sensors in Navigation. We have investigated here the low- field magnetic susceptibility of hole doped RE manganites with doping concentration x= 0.1 -0.5 . Our curves exhibit a PM – FM transition at a temperature T^* which increases as x increases. Well above T^*, the Curie – Weiss dependence of X_S (T) is observed. Upon lowering the temperature below T^*, anomalous behavior of X_S (T) which arises from the onset of magnetic ordering at T^* ~ 200 K is observed.

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