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A Variational Theory of Magnetic Susceptibility of Anderson Lattice Model: An Application to Colossal Magnetoresistance Manganites ($Re_{1-x}A_xMnO_3$)

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A R T I C L E I N F O

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A B S T R A C T

In the paper, a simple variational method for calculating the magnetic susceptibility(X_S) at different magnetic fields of rare earth manganites doped with alkaline earths namely $Re_{1-x}A_xMnO_3$ (where Re = La, Pr, Nd etc. and A= Ca, Sr, Ba etc.) which exhibit colossal magnetoresistance (CMR), metal-insulator transition and many other poorly understood phenomena. We have recently developed a two band (l-b) Anderson lattice model Hamiltonian along with (l-b) hybridization recently studied by us for manganites in the strong electron- lattice Jahn-Teller (JT) coupling regime an approach similar to the two-fluid models. Along with this variational method to study the zero field electrical resistivity and magnetic susceptibility of doped CMR manganites. In the present study, we find that the magnetic susceptibility (X_S) increases with increasing magnetic field *h*, *m* parameters and magnetic transition temperature T_c shifts towards lower temperature region. While at a particular value of h and m parameters, the maximum of X_S occurs at T_c (~ 100 K) followed by a sudden drop in X_S as we decrease the temperature, resembling with the key feature of many CMR compounds like $La_{1-x}Ba_xMnO_3$ with x=0.02-0.35 and $La_{0.67}Ca_{0.33}Mn_{0.9}Ta_{0.1}O_3$. This anomaly in X_S arises from the onset of magnetic ordering at 100K and vanishes on increasing *V* or J_H or doping *x* value. While it increases with increasing J_F value and the peak at low temperature becomes more sharpened.

K E Y W O R D S

A. Colossal magnetoresistance | D. Magnetic susceptibility | E. Variational method | E. Model parameters

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C I T A T I O N

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Introduction

Recently, there has been a surge interest (Schiffer *et al*., 1995) in the properties of rare earth manganites doped with alkaline earths namely $\boldsymbol{Re_{1-x}A_xMnO_3}$(where Re=La, Pr, Nd etc., and A= Ca, Sr, Ba *etc*.) which exhibit colossal magnetoresistance (CMR), metal- insulator transition and many other poorly understood phenomena. These systems are found to exhibit fascinating properties 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 (Millis, A., 1995) combining with the local Jahn-Teller distortions of Mn^{3+} ions (Uehara, M., 1999). Recently, it has been found that manganese oxides display a rich phase diagram and the coexistence of real-space variations of physical properties (Moreo, A., 1999). Percolation based on phase separation has been proposed to explain magneto transport properties in these systems (Mayr *et al*., 2001). In a review by Dagotto, Hotta and Moreo (2001), the phenomenon of phase separation has been discussed in manganites. In it, they explained theoretically the possibility of electronic phase separation between antiferromagnetic insulating and ferromagnetic metallic states. They have done extensive numerical simulations of several models but on rather small lattices and seen lots of instances of " phase separation" .In a recent review, Ramakrishnan *et al*. (2004) have presented a new theoretical model of coexisting localized JT polarons and broad band electrons for doped rare earth manganites $\boldsymbol{Re_{1-x}A_xMnO_3}$ and argued that it arises inevitably in the presence of orbital degeneracy and strong JT coupling and claimed that it explains a wide variety of characteristic properties of manganites. Some time ago, Panwar *et al*. (1989) have developed a variational method to study the ground state and thermodynamic properties of heavy fermion systems using Anderson lattice model. Recently, Panwar *et al*. had used this variational method in the study of the magneto transport properties like electrical resistivity and magnetic susceptibility etc. of doped CMR manganites over a fairly wide temperature range at zero and different magnetic fields (Panwar *et al*., 2014, Chaudhary *et al*., 2015, Panwar *et al*., 2015, 2017). In this paper, we continue to use this variational method to study the magnetic susceptibility of rare earth manganites doped with alkaline earths at different magnetic fields. In Section 2, we give the basic formulation for magnetic susceptibility $\boldsymbol{X}$s (T). In Section 3, we discuss our results and finally we conclude our findings in Section 4.

Basic Formulation

Model

We develop a theoretical model for the quantitative analysis of temperature dependent magnetic susceptibility $\boldsymbol{X}$s (T) at different magnetic field of manganites in the strong electron- lattice Jahn- Teller (JT) coupling regime. We start with a model Hamiltonian H_{lb} in the presence of magnetic field **H** which includes ℓ-b hybridization effects and thus can address the low temperature properties of manganites (*e.g.* resistivity ,Hall effect) (Ramakrishan *et al*., 2004 and Panwar *et al*., 2014) given by-

$$H_{lb} = -\sum_{\langle ij\rangle\sigma} \bar{t}_{ij} \left(b^{+}_{i\sigma} b_{j\sigma}\right) - \sum_{i\sigma} E_{jt}\, l^{+}_{i\sigma} l_{i\sigma} + U \sum_{i\sigma} n^{l}_{i\sigma}\, n^{b}_{i\sigma} - J_H \sum_i \boldsymbol{S_i} \cdot \boldsymbol{S_i} - J_F \sum_{\langle ij\rangle} \boldsymbol{S_i} \cdot \boldsymbol{S_j} - \sum_{k\sigma} V_K (l^{+}_{k\sigma} b_{k\sigma} + h.c.) - \mu B \sum_i \boldsymbol{S_i}.\mathbf{h}$$

(1)

and $\mathbf{h} = g\,\sigma\,\mu_B\,\mathbf{H}$.

Here $l^{+}_{i\sigma}$ creates the JT polaronic state of energy $-E_{jt}$ and spin σ localized at site i ($l_{i\sigma}$ is the corresponding destruction operator) and $b^{+}_{i\sigma}$ creates broad band electron having mean energy zero and nearest neighbour effective hopping amplitude $\bar{t}_{ij}$. $\boldsymbol{U}$ is the local Coulomb repulsion between ℓ - polarons and b-electrons of the same spin at a particular site i. $\boldsymbol{V_k}$ is the *l*-b hybridization between *l*-polarons and b-electrons of the same spin. $\boldsymbol{J_H}$ is the strong ferromagnetic Hund's rule coupling between the e_g spin $\vec{s_i}$ and the t_{2g} spin $\vec{S_i}$. $\mathbf{J_F}$ is the net effective ferromagnetic nearest neighbor exchange coupling between the $\boldsymbol{t_{2g}}$ core spins (S_i, S_j) and the last term of Eq. (1) denotes the interaction of the t_{2g} core spins with an external magnetic field **H**.

In the finite interaction U case, the modified variational wave-function (9) may be written as in k-space

$$\psi_{lb} = \prod_{k\sigma} [1 + A_{k\sigma} l^{+}_{k\sigma}\, b_{k\sigma}]\, |\Phi_d\rangle \qquad (2)$$

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

With this wave function, the variational parameter $A_{k\sigma}$ is given by

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

(3)

where

$\epsilon_k = \sum_{ij} \bar{t}_{ij} eik(Ri - Rj)$; Ri and Rj are the position vectors of i and j sites

(4)

and

$$\epsilon_{d\sigma} = \frac{U}{2}\, n^{b}_{\sigma} + \frac{J_H}{2}\, n^{l}_{\sigma} - E_{jt} - J_F\, \sigma\, m - \mathbf{h}$$

(5)

Here J_F involves the number of nearest neighbors and m is the magnetization per site given by

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

(6)

While the number of b-electrons and *l* - electrons are obtained by

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

(7)

and

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

(8)

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

$$f^{-}_{k\sigma} = 1/\exp(\beta(E^{-}_{k\sigma} - \mu) + 1)$$

(9)

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

The expression for $E^{-}_{k\sigma}$ is given by

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

(10)

Magnetic Susceptibility:

The static magnetic susceptibility 'χ_S ' is given by

$$\chi_S = g\,\sigma\,\mu_B \frac{\partial}{\partial B}\left(n^{l}{}_{\sigma} - n^{l}{}_{-\sigma}\right) B \text{ -->0}$$

(11)

where $n^{l}{}_{\sigma}$ and $n^{l}{}_{-\sigma}$ represent the number of ℓ - electrons of spin σ and -σ .

We obtain χ_S in the units of $(g\,\mu_B)^2$ as

$$\chi S(U, JF) = \sum_{k\sigma} \left(\frac{\chi 0(U)}{(1+\left(\frac{U}{2}\right)I3-(\left(\frac{U}{2}\right)+2JF)\chi 0(U)/2)}\right)$$

(12)

where $\chi_0(U) = I_1 + I_2$

(13)

$$I_1 = \frac{2A\ k\sigma\ Vk\ I3}{\sqrt{(\epsilon_k + \frac{U}{2} n^{l}{}_{\sigma} - \epsilon_{d_\sigma})^2 + 4V_k{}^2}}$$

(14)

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

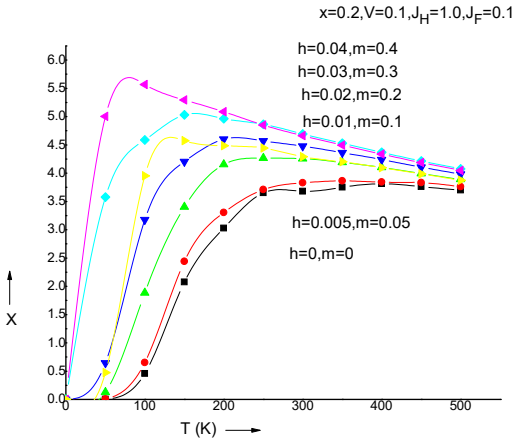
(15)

$$I_3 = \frac{A^2\ k\sigma}{(1+A^2\ k\sigma)} \beta \exp(\beta(E_{k\sigma}^{-} - \mu))(f_{k\sigma}^{-})^2$$

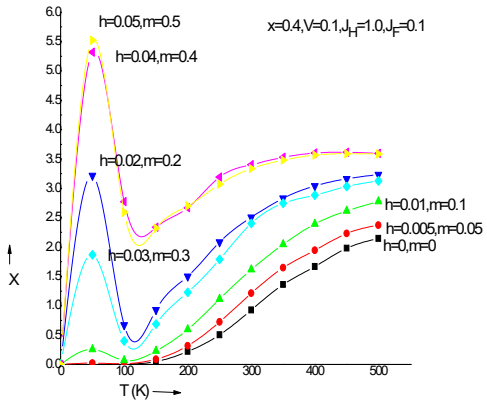
(16)

Results and Discussion

In our calculations, we have taken the unperturbed band of three dimensional solid represented by simple semi-circular 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, J_F= 0.1,V= 0.1and 0.2 ,ϵ_F = -0.238 eV (for x =0.3) and J_H = 1.0 and 2.0eV. Doping x is varied from 0.1 to 0.5.

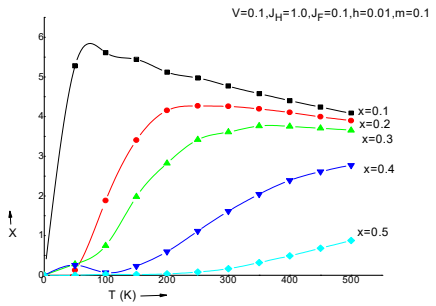


(a)

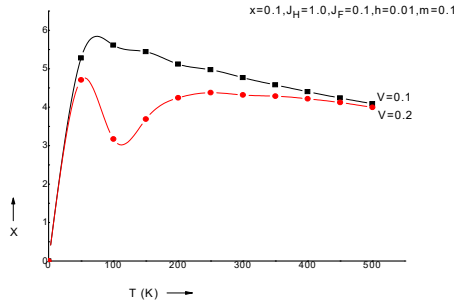


(b)

Fig. 1: Variation of magnetic susceptibility (X_S) with temperature T (K) for different values of h, m parameters at U=5.0,E_{jt}=0.5, V=0.1, J_H=1.0 and J_F=0.1 with (a) x=0.2 and (b) x=0.4



(a)



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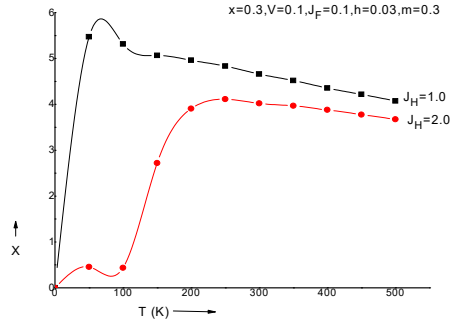

(c)

Fig 2 (a), (b) and (c): Variation of magnetic susceptibility (X_S) with temperature T (K) for different values of *x, V* and J_H parameters respectively at U=5.0,E_{jt}=0.5, h=0.01, 0.03and m=0.1, 0.3

In Fig.1, we have shown the temperature dependence of magnetic susceptibility (X_S) for different values of h and m parameters at a fixed value of *V*, J_H and J_F with (a) x=0.2 and (b) *x*=0.4.The parameter h is related to the physical field through h= g μ_B S_c H_{phys} / t. Using g=2, t=0.6 eV and S_c=3/2, we find that h= 0.01 corresponds to H_{phys} =15T (Millis *et al*., 1996) whereas the parameter m stands for magnetization per site given by Eq. (6) in Sec. 2.1.

In Fig. 1, for a particular value of h, m parameters, ***X*S** follows a Curie- Weiss behavior at high temperatures well above T_c. The Curie- Weiss law is violated with lowering of temperature. At low temperature it shows a peak at T_c (~100 K) followed by a sudden drop in ***X*S** as we decrease further the temperature resembling with the key feature of many CMR compounds likeLa_{1-x} Ba $_x$ MnO_3 (Laiho *et al*., 2005) with x=0.02-0.35 and $La_{0.67}$ $Ca_{0.33}$ Mn $_{0.9}$ $Ta_{0.1}$ O_3(Seetha *et al*., 2005). We also find that the ***X*S** increases with increasing magnetic field parameters h and m and magnetic transition temperature T_c shifts towards lower temperature region on increasing h and m parameters as evident from Fig. 1(a).

Fig. 1 shows the variation of magnetic susceptibility (***X*S**) with temperature in both the absence and in the presence of a magnetic field. For all cases, it is observed that with the application of a magnetic field, ***X*S** (T) values increase at low temperature. This indicates that spin ordering which occurs under magnetic field, increases the ***X*S** (T) of these systems. The observed behavior may be explained qualitatively by considering that with the application of a magnetic field, local ordering of magnetic spin occurs which causes the feeomagnetic metallic (FMM) state to suppress the paramagnetic insulating (PMI) state. As the spin ordering is more pronounced in the low temperature FM phase, the effect of magnetic field on ***X*S** (T) is more in this low temperature phase whereas in the PMI state

(T> Tc) the effect of magnetic field on the $\boldsymbol{X}_S$ (T) is comparatively smaller. The increase in $\boldsymbol{X}_S$ (T) due to applied field is maximum near the $\boldsymbol{X}_S$ (T) peak.

In Fig 2 (a),(b) and (c), we have shown the temperature dependence of magnetic susceptibility ($\boldsymbol{X}_S$) for different values of x, V and J_H parameters respectively at U=5.0,E_{jt}=0.5, h=0.01, 0.03and m=0.1, 0.3. Here we reach to the conclusion that the anomaly in $\boldsymbol{X}_S$ at low temperature (~100 K) becomes broader and shifts towards higher temperatures and even vanishes on increasing V or J_H or doping concentration x. This anomaly at 100 K in $\boldsymbol{X}_S$ arises from the onset of magnetic ordering and vanishes on increasing V, J_H or doping x value. The influence of doping on the magnetic susceptibility is clearly reflected. The paramagnetic to ferromagnetic transition temperature' Tc 'shifts to higher temperatures with the doping with a simultaneous decrease in the value of $\boldsymbol{X}_S$ (T). The ferromagnetic exchange interaction J_F enhances the $\boldsymbol{X}_S$ appreciably in the low temperature without affecting the temperature T_{max} at which the susceptibility shows a maximum (Chaudhary *et al*., 2015). We have also seen that the results of the simple model considered here are in qualitative agreement with the experimental results of a broad class of whole doped CMR manganites.

Conclusion

From the study of magnetic susceptibility (X_S) we tried to find the effect of magnetic field on the magnetic properties of CMR materials using the Variational method. In this study, the magnetic susceptibility ($\boldsymbol{X}_S$) increases with increasing magnetic field parameters h and m and magnetic transition temperature T_c shifts towards lower temperature region on increasing h and m parameters. We have also shown the temperature dependent magnetic susceptibility ($\boldsymbol{X}_S$) at different values of x, V and J_H parameters. We obtained that the anomaly in $\boldsymbol{X}_S$ at low temperature (~100 K) becomes broader and shifts towards higher temperatures and even vanishes on increasing V or J_H or doping concentration x. This anomaly at 100 K in $\boldsymbol{X}_S$ arises from the onset of magnetic ordering. In near future, we are going to study the effect of magnetic field on other finite temperature properties e.g. specific heat of these CMR materials.

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