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# Microscopic Model Study of the Role of Second Nearest Neighbour Spin-Spin Interaction in CMR Manganites

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**Abstract**: We report here a model study showing the effect of second nearest neighbour Heisenberg interaction on the interplay of the transverse spin fluctuation and band Jahn-Teller (JT) distortion. We have considered a double exchange (DE) model for an antiferromagnetically ordered manganite in the presence of JT interaction as an extra mechanism. The Hamiltonian is solved using Zubarev's Green's function technique to calculate the magnetic order parameters and lattice strain. The gap equations are solved numerically and self-consistently and the effect of first and second nearest neighbour Heisenberg interaction on the interplay of these order parameters is studied. Further conduction band electron density of states near Fermi level is studied. The results are in good agreement with experimental results.

**Keywords:** Colossal magneto-resistance; Jahn-Teller effect; Magnetization

**PACS:** 75.47.Gk; 71.70.Ej; 75.25.Dk

## 1. Introduction

Transition metal oxides such as hole doped rare earth manganites of general formula $R_{1-x}A_xMnO_3$ (R-rare earth ion and A-alkaline earth ion) exhibit many fascinating properties and phenomena due to the presence of a variety of electronic orders with spatially correlated charge, spin and orbital arrangements. After the discovery of colossal magneto resistive (CMR) property in $La_{2/3}$ $Ca_{1/3}$ $MnO_3$ thin film at 77K and 6T [1], study of rare-earth manganites has attracted scientists. The manganese ion is in a +3 state in the undoped rare-earth manganite ($RMnO_3$), whereas manganese ion in hole doped mangenites ($R_{1-x}A_xMnO_3$) is found in a mixed valence state ((1-*x*) of $Mn^{3+}$ and *x* of $Mn^{4+}$ states). The rich phase diagram of manganese oxides is mainly due to this mixed-valent

characteristic of Mn-ion. The CMR manganites exhibit spin, lattice and orbital orderings [2].

The undoped manganites show A-type antiferromagnetic (AFM) spin order which is accompanied by the orbital order (OO). The orbital ordering is strongly associated with lattice distortion due to the Jahn-Teller (JT) effect. In presence of JT active $Mn^{3+}$ ions, the local $MnO_6$ octahedra are elongated and shortened alternatively in the ab-plane and hence the doubly degenerate $e_g$ orbitals split to produce c-type orbital ordering. The JT distortion along with OO gives rise to ferromagnetism (FM) in the plane and AFM between the planes at low temperatures. As the hopping strength of electrons between the nearest neighbour (NN ) Mn ions is reduced for strong octahedral tilting, the possibility of hopping of electrons among the next nearest neighbour (NNN) Mn sites increases. Now both NN and NNN interactions play a role in stabilizing the magnetic structure.

Theoretical study of orthorhombic manganites by Kim & Min [3] suggests that the $t_{2g}$ hopping is essential for describing NN and NNN super exchange interaction and determining the sign of NNN coupling. They have found out that both $J_a$ & $J_b$ are positive. Microscopic model study and phase diagram of multiferroic perovskite manganites [4] shows that the strength of NN and NNN coupling are comparable and the NNN coupling plays a vital role in determining the magnetic properties. Further, the transport properties of manganites depend on the $e_g$ electron density of states (DOS). Presence of a pseudo gap (PG) of energy $2\Delta$ is observed in scanning tunneling microscopy study (STS) near Fermi level with finite DOS for $La_{0.7}Pb_{0.3}MnO_3$ system [5]. This PG is associated with localization of charges due to JT distortion. Similar PG is observed near the Fermi level spectrum of $Pr_{0.5}Sr_{0.5}MnO_3$ in resontent photoemission spectroscopy (RPES) study [6]. In the theoretical study of CMR Manganites using cluster dynamic mean-field theory [7] and Monte - carlo simulation [8], the PG is observed near Fermi level. The electron DOS for ferromagnetically ordered JT active CMR manganites in presence of hybridization between $e_g$ and $t_{2g}$ electrons [9], the DE model [10] and the charge ordered (CO) CMR manganites with AFM ordering [11] is studied by using Zubarev's Green's function technique.

In the present communication, we study the effect of second nearest neighbour Heisenberg interaction theoretically for the antiferromagnetically ordered manganites in presence of band JT distortion in the conduction band and double exchange (DE) interaction between the $e_g$ and $t_{2g}$ electron spins. Further we study the effect of NNN Heisenberg coupling on the $e_g$ electron DOS. The

formalism is described in section 2 and the results and discussion are presented in section 3 and finally the conclusion is given in section 4.

## 2. Formalism

The Kubo-Ohata type [12] double exchange spin interaction and the Heisenberg type spin-spin interaction in core electrons are considered within mean-field approximation. The orbital ordering due to Jahn-Teller distortion splits the $e_g$ electron bands into two for two orbitals $\alpha = 1,2$. Based on our earlier model [13] the total mean-field Hamiltonian for the Manganite system is written as

$$H = \sum_{\alpha,k,\sigma} \epsilon_{\alpha,k,\sigma}\, C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma} \; - \frac{1}{2} J_{DE} \sum_{\alpha,k,\sigma} < S^{d} > C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,-\sigma} - \sum_{k,\sigma} J_{DH}\, d^{\dagger}_{k,\sigma} d_{k,-\sigma} + \sum_{k,\sigma} \epsilon_{d,\sigma}\, d^{\dagger}_{k,\sigma} d_{k,\sigma} + \frac{1}{2} Ce^{2} \qquad (1)$$

Here

$$\epsilon_{\alpha,k,\sigma} = \sum_{\alpha,k,\sigma} (\epsilon_k - (-1)^{\alpha} Ge - \mu - B\sigma$$

with the conduction band dispersion energy, $\epsilon_k = -2t_1\left(cosk_x + cosk_y\right) - 4t_2 cosk_x cosk_y$ , $t_1$ and $t_2$ being the first and second nearest neighbour hopping integrals. G is the static band JT coupling strength. $\alpha = 1,2$ for the two JT split bands. The chemical potential and external magnetic field are represented by μ and B ($= B_{ext}\mu_B g_L$) respectively. The value of $\sigma$=+1 for up-spin and -1 for down-spin electrons. The second term in the Hamiltonian represents Kuboo-Ohata [12] type double exchange (DE) interaction among the on-site spins of $e_g$ and $t_{2g}$ band electrons. $J_{DE}$ is the DE coupling constant and $< S^{d} >$ represents average transverse spin fluctuation in $t_{2g}$ band electrons. In the third term $J_{DH} = \frac{1}{2}(J_{DE} < s^{c} > + J_H < S^{d} >)$, where $J_H$ is the strength of Heisenberg interaction among the intersite spins of same core band. Due to double exchange interaction AFM is induced in conduction band and the induced AFM spin fluctuation is represented by $< s^{c} >$. We have considered both NN and NNN interaction among the core spins and hence Heigenberg coupling is written as $J_H = J_1(cosk_x + cosk_y) + 2J_2 cosk_x \cos k_y$. The kinetic energy of core electrons is given in fourth term of equation (1) with $\epsilon_{d,\sigma} = \epsilon_d - B\sigma$, $\epsilon_d$ being the position of core level with respect to Fermi level. The terms $C^{\dagger}_{k,\sigma}\left(C_{k,\sigma}\right)$ and $d^{\dagger}_{k,\sigma}(d_{k,\sigma})$ represent creation (annihilation) operators of $e_g$ and $t_{2g}$ band electrons

respectively. The last term represents the lattice energy with lattice constant C and lattice strain e in conduction band. The model Hamiltonian is solved using Zubarev's Green's function technique [14] and the coupled Green's functions for $e_g$ and $t_{2g}$ electrons are calculated. From the correlation functions obtained, the magnetic order parameters and lattice strain are calculated. The lattice strain is defined as

$$e = \frac{-G}{C} \sum_{\alpha,k,\sigma} (-1)^{\alpha} \bar{n}^{c}_{\alpha,k,\sigma} \tag{2}$$

where $\bar{n}^{c}_{\alpha,k,\sigma} = \frac{1}{2}[f(\beta\omega_{1,\alpha,\sigma}) + f(\beta\omega_{2,\alpha,\sigma})]$ is the average $e_g$ electron occupation number with quasiparticle energy of $e_g$ electrons $\omega_{i,\alpha,\sigma} = \epsilon_{\alpha,k,\sigma} - (-1)^{i} \frac{J_{DE}}{2} < S^{d} >$ , i=1,2. The induced average transverse spin fluctuation in conduction band

$$< s^{c} > = \sum_{\alpha,k,\sigma} < C^{\dagger}_{\alpha,k,\sigma} \; C_{\alpha,k,-\sigma} >$$

$$= -\frac{1}{2} \sum_{\alpha,k,\sigma} [f(\beta\omega_{1,\alpha,\sigma}) - f(\beta\omega_{2,\alpha,\sigma})] \tag{3}$$

and the average transverse spin fluctuation in core $t_{2g}$ band

$$< S^{d} > = \sum_{k,\sigma} < d^{\dagger}_{k,\sigma} \, d_{k,-\sigma} > = -\frac{1}{2} \sum_{k,\sigma} [f(\beta\omega_3) - f(\beta\omega_4)] \tag{4}$$

where the core electron quasiparticle energies are , $\omega_{3,4} = \epsilon_{d,\sigma} \pm J_{DH}$. The physical parameters are scaled with respect to the nearest neighbour hopping integral $t_1$=0.25eV =2500K. The reduced parameters are NNN hopping integral $t_2 = \frac{\widetilde{t_2}}{t_1}$, DE coupling $g_1 = \frac{J_{DE}}{t_1}$ , static JT coupling g=$\frac{G}{t_1}$ , the $t_{2g}$ NN-AFM spin coupling $g_{s1} = \frac{J_1}{t_1}$ , the $t_{2g}$ NNN-AFM spin coupling $g_{s2} = \frac{J_2}{t_1}$ , reduced lattice constant $c_1 = \frac{C}{t_1}$ , reduced temperature t = $\frac{K_B T}{t_1}$ , reduced band energy w = $\frac{\omega}{t_1}$ , reduced chemical potential $u_m = \frac{\mu}{t_1}$, and spectral width ee = $\frac{\eta}{t_1}$ .

## 3. Results and Discussion

The orbital ordering (OO) and spin ordering (SO) gap equations given in equations (2), (3) and (4) are solved numerically and self-consistently. For a set of constant coupling parameters, the static JT coupling, g = 0.2305, the DE

coupling $g_1$ =0.5, the NNN hopping integral $t_2$ = 1.75, the NN Heisenberg coupling $g_{s1}$= 0.68 and the NNN Heisenberg coupling $g_{s2}$= 0.475, the temperature dependent lattice strain (e), average transverse spin fluctuation in core band $\langle S^d \rangle$ and induced transverse spin fluctuation in conduction band $\langle s^c \rangle$ are plotted in figure 1 (solid line). The temperature dependent $\langle S^d \rangle$ graph exhibits mean- field like behaviour with AFM transition temperature at $t_N$ = 0.06 ($T_N \simeq$150K). The induced AFM spin fluctuation $\langle s^c \rangle$ also exhibits similar behaviour as that of $\langle S^d \rangle$ with the same Neel temperature. The temperature dependent lattice strain (e) produced in the $e_g$ band due to the lattice distortion in presence of static band JT distortion should also exhibit mean-field like behavior. As seen from figure 1, the strain is suppressed slightly from the mean-field nature in the spin ordering temperature region ( $t < t_N$) and the mean-field nature persists above $t_N$.

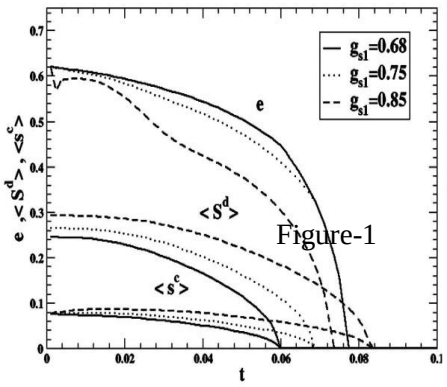


Figure-1

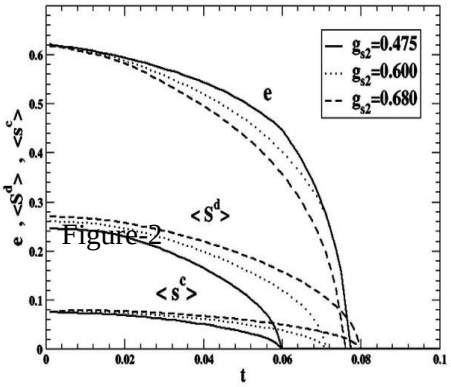


Figure-2

**Figure- 1** **Figure-2**

**Fig. 1.** Self-consistency plot of temperature dependent of e, $\langle s^c \rangle$ and $\langle S^d \rangle$ for fixed values of $g_1$=0.5, g=0.2305, $g_{s2}$=0.475, $c_1$=0.01, $t_2$=1.75 and different values $g_{s1}$=0.68, 0.75, 0.85. **Fig. 2.** Self-consistency plot of temperature dependent of e, $\langle s^c \rangle$ and $\langle S^d \rangle$ for fixed values of g=0.2305, $g_1$=0.5, $g_{s1}$=0.68, $c_1$=0.01, $t_2$=1.75 and different values $g_{s2}$=0.475, 0.600, 0.680.

The OO transition temperature is set at $t_{00}$ = 0.0775 ($T_{00} \simeq$ 194K). There are experimental evidences where $T_N < T_{00}$ for rare-earth manganites. For example, $T_{oo} \simeq$ 204K and $T_N \simeq$ 178K for $La_{0.2}Ca_{0.8}MnO_3$ system [15] and $T_{co} \simeq$230K and $T_N \simeq$152K for $La_{0.25}Ca_{0.75}MnO_3$ [16]. As the value of $g_{s1}$ is increased from 0.68 to 0.75 keeping other parameters constant, the AFM spin fluctuations in $t_{2g}$ and $e_g$ band are enhanced throughout the temperature range as well as the Neel temperature is increased from 0.06 to 0.068. Increase of the magnetic ordering in the $e_g$ band suppresses the orbital ordering in the interplay temperature region.

This is reflected by suppression of lattice strain (e) up to t=0.068 (shown by dotted line). Outside the interplay region the lattice strain is not altered. The OO temperature also remains unchanged. As the value of $g_{s1}$ is further increased to 0.85, the Neel temperature increases to 0.084. In this case, the value of e is suppressed throughout the temperature range as well as the $t_{oo}$ is shifted to a lower value showing a strong interplay between the magnetic and orbital ordering.

The effect of NNN coupling ($g_{s2}$) on the interplay of e, $\langle S^d \rangle$ and $\langle s^c \rangle$ is studied keeping the NN coupling ($g_{s1}$) constant at 0.68, which is shown in figure 2. With increase of $g_{s2}$ from 0.475 to 0.68, the AFM order parameters $\langle S^d \rangle$ and $\langle s^c \rangle$ increase. This increase in spin ordering is more near AFM transition temperature and less at lower temperatures. Also the Neel temperature is increased from 0.06 to 0.08. The involvement of more and more $e_g$ spins in AFM ordering decreases the lattice strain (e) in the interplay region. For $g_{s2}$=0.68, where $t_N > t_{oo}$, the OO temperature also lowers down. As observed from our model study, the NN and NNN coupling parameters are comparable with each other. So we cannot ignore the effect of second nearest neighbor interaction. Similar results are obtained in the microscopic model study of multiferroic manganites [4].

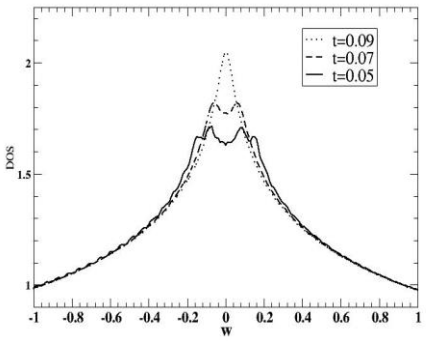


**Figure- 3**

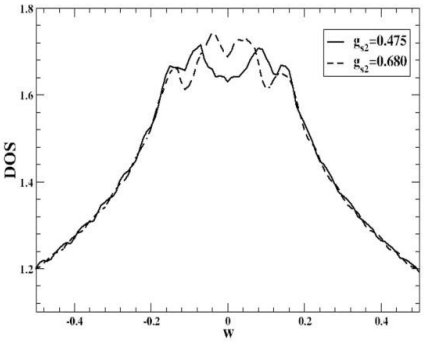


**Figure-4**

**Fig. 3.** The plot of density of states (DOS) versus band energy (w) for different values t=0.09, 0.07, 0.05, and fixed values of $g_{s2}$=0.475, g=0.2305, $g_1$=0.5, ee=0.015,$u_m$=0.57.
**Fig. 4.** The plot of density states (DOS) versus band energy (w) for different values $g_{s2}$=0.475, 0.680 and fixed values of g=0.2305, $g_1$=0.5, ee=0.015, $u_m$=0.57.

We have calculated the $e_g$ electron density of states (DOS) from the imaginary part of the $e_g$ electron Green's function $G_1(\alpha, k,\omega)$ using the formula $DOS = -2\pi \, \mathrm{Im} \, G_1(\alpha,k,\omega + i\eta)$, where $\eta$ is the small spectral width. Figure 3. shows the variation of $e_g$ electron DOS with respect to band energy at different

temperatures. At a higher temperature t = 0.09, in the paramagnetic (PM) phase, in absence of orbital ordering ($<S^d>$= 0 and e = 0), a single peak is obtained at w = 0 (at Fermi energy). The peak like structure appears at w=0 due to the small spectral width. As the temperature is lowered down to t = 0. 07, where only the orbital ordered state is present and the AFM spin ordering is absent ($<S^d>$= 0), the DOS peak obtained near Fermi level splits into two with a U-shaped energy gap. This splitting of the peak is associated with orbital ordering. The U-shaped insulating gap observed near Fermi level is due to the JT distortion. The energy gap between the two peaks $\simeq$0.13, which is approximately same as JT energy gap (2ge=2×0.2305×0.2975=0.137). Though there is a dip produced in the electron DOS near Fermi level, the DOS of $e_g$ state is not equal to zero. Such parabolic gaps are observed in the STS study of $La_{0.7}Pb_{0.3}MnO_3$ due to JT distortion [5] and RPES study of $Pr_{0.5}Sr_{0.5}MnO_3$ due to charge ordering [6]. Similar PG is observed near Fermi level in theoretical electron DOS study for manganites [7,8]. When we go to a still lower temperature t = 0. 05, in the co-existing phase of orbital ordering and spin ordering (e = 0.5046 and $<S^d>$= 0.1152), each of the two JT split peaks further split into two because of spin ordering. The four peaks are obtained at energies $\pm(ge+\frac{1}{2}g_1<S^d>)$ = ±0.14511 and $\pm(ge -\frac{1}{2}g_1<S^d>)$ = ±0.08751. The authors have reported that they could not observe multiple peak structures in STS experiments due to lack of unavailability of high resolving power photoemission spectroscopes [5].

The effect of second nearest neighbour interaction on the $e_g$ electron DOS is shown in figure 4. The DOS plots are drawn at a temperature t = 0.05, in the co – existing phase of OO and SO for two different values of NNN coupling $g_{s2}$=0.475 and 0.680. It is observed that as the value of $g_{s2}$ increases from 0.475 to 0.68, both the inner and outer peaks come closer to each other. Further the increase in spectral height and decrease in the gap between the inner peaks indicates an increase in electron DOS near Fermi level. The combined gap due to both OO and SO ($2(ge +\frac{1}{2}g_1<S^d>)$), which is the energy difference between the two outer gaps, also decreases indicating increase of metallic behavior of the system. An increase in NNN hopping leads to a decrease in JT distortion in orthorhombic perovskite manganites [3].

## 4. Conclusion

In the microscopic model study of JT active antiferromagnetically ordered manganite systems in presence of DE interaction, we find out that the second nearest neighbour interaction plays a vital role in the interplay of AFM order and

lattice strain. We observe a parabolic PG near Fermi level in the $e_g$ electron DOS which is associated with the JT insulating gap. Such PG are observed in the experimental studies of $La_{0.7}Pb_{0.3}MnO_3$ [5] and $Pr_{0.5}Sr_{0.5}MnO_3$ [6] systems.

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