Model of the Interplay of Band J-T Effect with Magnetic Order Mediated by Exchange Interaction

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A model calculation is presented with the aim to study the interplay between magnetic and structural transitions. The model consists of an orbitally doubly degenerate conduction band and a periodic array of local moments. The band electrons interact with the local spins via the s-f interaction. The interaction of the band electrons with phonons is introduced by including band Jahn-Teller (J-T) interaction. The model Hamiltonian, including the above terms, is solved for the single particle Greens function. In doing this an ansatz for selfenergy of electrons, which was developed earlier has been utilized. The quasiparticle density of states (QDOS) and hence the KLM are extended and form a doubly degenerate band. Out of the four d-electrons, three occupy the $t_{2g}$-states and the remaining electron is in the $e_g$-band. The $Mn$ ion is in $Mn^{3+}$ state. Due to the large Hund’s rule coupling in this system, the three $t_{2g}$-electrons have their spins oriented parallel to each other making a localized spin of $S = 3/2$. This is again strongly Hund’s rule coupled to the $e_g$-electron. Since the $e_g$ is an extended state, the electron in this state can hop from lattice site to lattice site. The hopping combined with Hund’s rule coupling is responsible for the long range magnetic order that exists in this system. This is known in literature as the double exchange mechanism. Another way of looking at the system is that there is a localized spin at each lattice site and the band electrons interact with these spins via an intraatomic exchange interaction. This is known in literature as the Kondo-lattice model (KLM). Similar scenario also prevails in Heusler alloy like $Ni_2MnGa$, where $Mn$ possesses localized moment and the magnetic properties of the host system can be understood from the KLM with the carrier concentration ($e_g$-electrons) of one per $Mn$ atom. When the trivalent rare-earth ion is partly replaced by divalent ions like $Ca$, $Sr$ or $Ba$, the material exhibits CMR properties undergoing transition from the paramagnetic-insulator state to the ferromagnetic-metal one. The localized spin of $S = 3/2$ is retained but there is a decrease in the electron concentration in the $e_g$-band. The carrier concentration is less than one per atom. While studying the CMR and the associated magnetic and insulator-metal transitions, it was realized that KLM alone is not sufficient to understand the physics of
manganites. It is now accepted that electron-phonon interaction plays an important role in the magnetic phase, which is driven by the lattice distortions (phonons) in the KLM model. This interplay has been reported in various manganites such as La$_2$MnO$_4$, Nd$_2$MnO$_4$, and Pr$_2$MnO$_4$. Obviously, this introduces J-T effect which lifts the degeneracy of the $e_g$-band. The spontaneous distortion associated with the J-T effect exists when the lowering of the band energy is more than the increase in elastic energy due to the strain. The simplest way to describe the band J-T effect is to incorporate the interaction of the $e_g$-electrons with the distortions (phonons) in the KLM model. When this is done, it is pertinent to examine the interplay between the J-T effect and magnetism in the model system. There are experimental results supporting such an interplay in manganites and in Heusler alloys. It was observed that the J-T distortion in (La - Y)CaMnO$_3$ and (La, Pr, Ca)MnO$_3$ is reduced in ferromagnetic state. The suppression of the J-T distortion in Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ ferromagnetic state has been reported. The coexistence of the J-T distorted phase and the ferromagnetic phase have been reported in Rh$_2$CoS$_3$. J-T splitting exists in these systems below the transition temperature $T_C$.

Therefore, in order to study the interplay of structural and magnetic transitions, we consider a model where the band electrons, which are approximated to be s-electrons in doubly degenerate extended states interact intratotically with a periodic array of localized spins (a Kondo lattice). We provide for the spontaneous lifting of the degeneracy of the band states by including a band J-T interaction. The lifting of the degeneracy is signalled by the appearance of strain. The presence of the long range magnetic order is characterized by the nonzero value of the magnetization. We study, selfconsistently, the strain as a function of the J-T coupling constant for different carrier ($e_g$-electron) concentrations and the dependence of the strain on temperature. In the latter case, the magnetization of the local moments which is caused by the exchange interaction between the $e_g$- and $t_{2g}$-electrons (Kondo interaction) determines, decissively, the temperature dependence of the strain.

II. MODEL HAMILTONIAN AND ITS APPROXIMATE SOLUTION

The $e_g$-electrons moving in the doubly degenerate band are described by

$$H_s = \sum_{\alpha=1}^{2} \sum_{ij\sigma} (T_{ij} - \mu \delta_{ij}) c_{\alpha i\sigma}^\dagger c_{\alpha j\sigma}$$

$$= \sum_{\alpha k\sigma} (\epsilon(k) - \mu) c_{\alpha k\sigma}^\dagger c_{\alpha k\sigma}$$

(1)

$T_{ij}$ is the hopping integral for hopping of the electrons from lattice site $i$ to $j$. $c_{\alpha i\sigma}^\dagger (c_{\alpha i\sigma})$ is the creation (annihilation) operator for an electron in the $\alpha$-state on the lattice site $i$ with spin $\sigma$. $\alpha = 1, 2$ is the band index. $\mu$ is the chemical potential. $\epsilon(k)$ is the band energy related to $T_{ij}$ by

$$T_{ij} = \frac{1}{N} \sum_k \epsilon(k) e^{i k \cdot (R_i - R_j)}$$

(2)

The band electrons interact with the localized spins via the intraatomic exchange interaction of the coupling strength $J$ and this is described by

$$H_{sf} = -J \sum_{j, \alpha} \sigma_{\alpha j} \cdot S_j$$

$$= -\frac{1}{2} J \sum_{\alpha, j\sigma} (z_\sigma S_j^z n_{\alpha j\sigma} + S_j^{-\sigma} c_{\alpha j-\sigma}^\dagger c_{\alpha j\sigma})$$

(3)

$\sigma$ is the spin of the band electron and $S$ is the localized spin (total spin of the three $t_{2g}$ electrons). $n_{\alpha j\sigma}$ is the number operator for the electron in the state $\alpha$ at the lattice site $j$ with spin $\sigma$. At the outset itself we assume a ferromagnetic interaction ($J > 0$). $z_\sigma$ is a sign factor, $z_\sigma = \delta_{\sigma - 1} - \delta_{\sigma + 1}$ and $S_j^z = S_j^+ + i z_\sigma S_j^-$. The electron density in the degenerate band couples to the static elastic strain through the J-T interaction. In the case of a tetragonal distortion, this interaction is described by

$$H_{JT} = G e \sum_{k, \sigma} (n_{1k\sigma} - n_{2k\sigma}) = G e \sum_{i\sigma} (n_{1i\sigma} - n_{2i\sigma})$$

(4)

$G$ is the strength of the J-T coupling and $e$ is the lattice strain given by

$$e = \frac{G}{NC_0} \sum_{i\sigma} (n_{1i\sigma} - n_{2i\sigma})$$

(5)

where $C_0$ is the elastic constant. It is clear that $H_{JT}$ tries to create a difference in the occupation of the two degenerate bands. The difference in occupation leads to the building up of the strain. Thus, under suitable conditions, there is a spontaneous splitting of the bands and building up of strain which indicates a structural transition. The building up of the strain however leads to an increase in the lattice elastic energy which is given by

$$H_L = \frac{1}{2} N C_0 e^2$$

(6)

Where $N$ is the total number of atoms. Since this term is a c-number and we are not looking for the ground state whose energy has to be minimum, we leave this term out of our consideration. Then the Hamiltonian of the model system we are considering is

$$H = H_s + H_{sf} + H_{JT}$$

(7)

The model Hamiltonian Eq(7) obviously cannot be solved exactly. However, in an earlier work, we have proposed,
for the Hamiltonian without the J-T term, an approximation scheme, which is reliable in the limit of low carrier concentration. We will exploit that scheme in solving the present model. Firstly, without resorting to any approximation, we can absorb the J-T term into $H_s$ by modifying the band energies for the two bands as

$$\epsilon_\alpha(k) = \epsilon(k) + (-1)^\alpha Ge. \quad (8)$$

Then we have

$$H_s = \sum_{\alpha k \sigma} (\epsilon_\alpha(k) - \mu) c_{\alpha k \sigma}^\dagger c_{\alpha k \sigma}$$

and the total Hamiltonian is given by

$$H = H_s + H_{sf}. \quad (9)$$

In order to calculate the strain (caused by the structural transition), one has to calculate the one-electron Greens function

$$G_{\alpha k \sigma}(E) = \langle \langle c_{\alpha k \sigma}^\dagger c_{\alpha k \sigma} \rangle \rangle_E$$

$$= \frac{1}{E - \epsilon_\alpha(k) - \Sigma_{\alpha \sigma}(E)}.$$ \quad (11)

That means, one has to calculate the selfenergy $\Sigma_{\alpha \sigma}(E)$ of the electron in the presence of $H_{sf}$. There are some exact results available for the selfenergy in certain limiting cases, namely, the zero band width limit for all temperatures\textsuperscript{19} and the finite band width but ferromagnetic saturation ($T = 0$) limit\textsuperscript{20,21}. In addition, using Mori formalism, the result for the second order perturbation theory is also available\textsuperscript{22}. We propose an ansatz for the self energy that reproduces the known limiting results and in addition, satisfies the strong coupling limit. This can be taken care of by making a high energy expansion and evaluating the first four spectral moments\textsuperscript{22}. We thus have a selfenergy which fulfills a) the zero band width limit for all temperatures and coupling strengths, b) $T = 0$ limit for all band widths and coupling constants, c) the weak coupling limit ($O(J^2)$) and d) has the correct high energy behavior. The ansatz for $\Sigma_{\alpha \sigma}(E)$ is given by

$$\Sigma_{\alpha \sigma}(E) = -\frac{1}{2}J_m^\sigma + \frac{1}{4} J^2 \frac{a_\sigma G_{\alpha 0}(E - \frac{1}{2}J_m^\sigma)}{1 - b_\sigma G_{\alpha 0}(E - \frac{1}{2}J_m^\sigma)}. \quad (12)$$

Here

$$G_{\alpha 0}(E) = \frac{1}{N} \sum_k G_{\alpha k}(E) = \frac{1}{N} \sum_k \frac{1}{E - \epsilon_\alpha(k)}.$$ \quad (13)

$m^\sigma = z^\sigma (S^z)$. The ansatz assumes a $k$-independent selfenergy. As $H_{sf}$ is a local interaction, the energy dependence of the selfenergy is the deciding factor in relation to the electron density of states. The parameters $a_\sigma$ and $b_\sigma$ and are fixed by rigorous high energy expansions to fulfill the first four spectral moments:

$$a_\sigma = S(S+1) - m^\sigma (m^\sigma + 1) \quad b_\sigma = b_{-\sigma} = \frac{1}{2} J.$$ \quad (14)

It should be mentioned that this ansatz is valid only in the limit of low carrier density. Since we are interested in simulating systems with low carrier density, it is justified to use the above ansatz. From $G_{\alpha k \sigma}(E)$ one can obtain the spectral density $S_{\alpha k \sigma}(E)$ and the density of states $\rho_{\alpha \sigma}(E)$ from the well known relations

$$S_{\alpha k \sigma}(E) = -\frac{1}{\pi} Im G_{\alpha k \sigma}(E) \quad (15)$$

$$\rho_{\alpha \sigma}(E) = \frac{1}{N} \sum_k S_{\alpha k \sigma}(E) \quad (16)$$

From the knowledge of the density of states, the expectation values can be evaluated:

$$\langle n_{\alpha \sigma} \rangle = \int dE f_-(E) \rho_{\alpha \sigma}(E) \quad (17)$$

Where $f_-(E) = 1/(1 + e^\beta E)$ is the Fermi function with $\beta = 1/kT$. The chemical potential $\mu$ is fixed by the constraint

$$n = \sum_{\alpha \sigma} \langle n_{\alpha \sigma} \rangle = \text{constant}. \quad (18)$$

For a given set of the model parameters $n, J, S$ and $G$ and for a fixed $T$, the occupancies of both lower and upper sub-bands for each spin directions is computed self-consistently. After having the self consistent solution, the lattice strain

$$e = \frac{G}{C_0} \sum_\sigma (\langle n_{1\sigma} \rangle - \langle n_{2\sigma} \rangle)$$ \quad (19)

is calculated. The average occupation of $e_g$-orbitals $\langle n_{\alpha \sigma} \rangle$ can be numerically obtained using a model density of states for the "free" $e_g$-band:

$$\rho_0(E) = A \sqrt{1 - \frac{E}{D}} \ln \left| \frac{D^2}{E^2} \right|$$ \quad (20)

Where $A$ is a normalization constant and $D$ is half the width of free Bloch band. In order to calculate $\langle n_{\alpha \sigma} \rangle$, we require $\langle S^z \rangle$, since this enters into the selfenergy. The local moment system is described within the mean field approximation and is represented by Brillouin function. The effective field seen by the local moment is determined by mutual exchange interaction which fixes $T_C$. Numerical results are given where all the energy parameters are normalized in terms of the free bandwidth $(2D)$. Ideally, one should get the magnetization $\langle S^z \rangle$ self-consistently out of the calculation. However, it is a very involved problem. Therefore, we treat it as a parameter and obtain its value at any temperature from the Brillouin function assuming a value for $T_C$.

Using the above theory, the results for the density of states and the strain are presented in the next section.
III. RESULTS AND DISCUSSION

First we consider the $T = 0$ case for two extreme situations of the local magnetization, namely, the paramagnetic ($\langle S^z \rangle = 0$) and the saturated ferromagnetic ($\langle S^z \rangle = S$) state. In Fig.1 we have plotted the strain as function of the J-T coupling constant $G$. We find that unless the value of $G$ exceeds a critical value, there is no spontaneous splitting of the orbitally degenerate bands. The splitting of the bands takes place and the lower of the split bands is occupied more by the electrons than the upper one in order to lower energy. This is energetically favoured only for a sufficiently large $G$. When $G$ is further increased, the lower of the split bands is more populated and therefore the strain increases as shown in the figure. Though not shown in the figure, the critical value of $G$ also depends on the carrier concentration. Larger is $n$, smaller is the critical value of $G$. Another feature which is displayed in Fig.1, is the role of the magnetization $\langle S^z \rangle$ at $G = 0$. We find that smaller is the critical value of $G$, larger is the role of the magnetization $\langle S^z \rangle$ at $G = 0$. The structural transition temperature $T_s$ is the temperature at which the strain goes to zero. We choose the parameters such that $T_s = 800K$. Then we study the effect of magnetization on strain by varying $T_C$ such that i) $T_C < T_s$, ii) $T_C \approx T_s$ and iii) $T_C > T_s$. The Fig. 2 displays the T-dependence of the strain(curves a, b c d and e) and the magnetization (which is a Brillouin function for a given $T_C$) for different $T_C$'s (curves 1, 2, 3, and 4). Whatever is $T_C$, at $T = 0$, $\langle S^z \rangle = S$. Therefore, the effect of the magnetization on the strain is independent of $T_C$ and leads to a maximum decrease of strain. As $T_C$ increases, the strain goes up with increasing $T$ and the rate of increase is higher for lower $T_C$ so long as $T_C < T_s$. Therefore, there always appears a peak in the curves. When $T_C \approx T_s$, the peak is very faint and the strain becomes very small well before $T_s$ is reached. For $T_C > T_s$, the strain is nonzero only when $T$ is much lower than $T_C$ (curve e of Fig.2).

It is clear that at $T = 0$, the presence of magnetization causes a redistribution of electrons between the orbital levels by creating a population difference between the...
spin levels. Such redistribution is the cause of suppression of strain. As $T$ increases, the spin level occupancies tend to equalize and that polarises the orbital levels further resulting in the increase in the strain compared to its $T = 0$ value. Since for smaller $T_c$, the magnetization decreases faster with increasing $T$, the increase in strain is also faster. For $T_c > T_s$, due to the choice of parameters, as expected, there is no strain between $T_s < T < T_c$. When $T$ is much lower than $T_c$, the occupancy of the spin levels is stabilized and the system can lower energy by further redistribution of electrons between the orbital states. That is why the strain is finite for $T$ much less than $T_c$ (curve e of Fig. 2). The results obtained are interpreted with the help of the quasiparticle density of states (QDOS). Before discussing the results of the full problem, to fix up a reference for further discussion, we want to present in Fig.3, the QDOS for the case of a pure KLM, that is, for the case of $G = 0$. The QDOS consists of two subbands for each spin direction separated by an energy of the order of $\frac{1}{2}J(2S + 1)$. The separation of the bands is independent of $T$ but the spectral weights of these subbands, however, depend on $T$ through the value of $\langle S^z \rangle$. For example, at $T = 0$ ($\langle S^z \rangle = S$), the spectral weight of the upper subband for $\uparrow$-states is zero. The reason for this is easy to understand. At $T = 0$, the local moment system is saturated. Therefore, for an $\uparrow$-electron there is no chance to spin-flip by involving a corresponding spin-flip of the local moment system. That means, at $T = 0$, as far as the $\uparrow$-electron is concerned, only the Ising part of $H_{sf}$ operates resulting simply in a rigid shift of QDOS. As we see from Fig.3, the spectral weight of the $\downarrow$-states in the lower sub band is finite. This is because, for a $\downarrow$-electron, even at $T = 0$, spin-flip is possible. Furthermore, when a $\downarrow$-electron flips its spin, it lands as an $\uparrow$-electron. Therefore the nonzero QDOS of the $\downarrow$-electron should be in the same energy region as that of $\uparrow$-electron. For a $\downarrow$-electron there is another possibility. It can have repeated magnon emission and absorption. That is, in a sense, it propagates in the lattice dressed by a cloud of magnons. This is a stable quasiparticle, which we call the magnetic polaron. Obviously, at $T = 0$, there is no possibility of magnetic polaron for $\uparrow$-electron. As $T$ increases ($\langle S^z \rangle$ decreases), the spin flip processes are allowed for both the spin directions and therefore the spectral weights in both the subbands are nonzero for both the spin directions. At $T = T_c$ ($\langle S^z \rangle = 0$), the spectral weights of $\uparrow$- and $\downarrow$-states in the two subbands become equal as it should be. We note the asymmetry with respect to the centre of the free band. This originates from the renormalization of the atomic levels by the s-f interaction.

Now we consider the further splitting of these bands due to the J-T effect. That is, when the degeneracy of the $e_g$ band is lifted due to the J-T effect, each of the subbands of Fig.3 for each spin direction discussed above is again split into two as shown in Fig.4. Noting the position of the chemical potential, $n = 0.7$, $J = 1$, $S = 3/2$, $C_0 = 1$, ($\langle S^z \rangle = S$ and $T = 0$).

![FIG. 3: Quasiparticle density of states as a function of energy for various values of localized magnetization ($S^z$) in the absence of Jahn-Teller distortion i.e for $G = 0$. Full line for spin up and dotted line for spin down. $J = 1$ and $S = 3/2$](image1)

![FIG. 4: Quasiparticle density of states (for lower sub-band($\alpha = 1$) in the positive half of the frame and for upper sub-band($\alpha = 2$) in the negative half of the frame) as a function of energy for various values of electron-phonon coupling constant $G$. Full line for spin up and dotted line for spin down. Thin vertical line shows the position of the chemical potential, $n = 0.7$. $J = 1$, $S = 3/2$, $C_0 = 1$, ($\langle S^z \rangle = S$ and $T = 0$).](image2)
on the coupling constant $G$ and the influence of $\langle S^z \rangle$. In Fig.4, we take the case of saturation ($\langle S^z \rangle = S$). When $G$ is less than the critical value, the two $e_g$ bands are degenerate. As $G$ approaches the critical value of 0.42, there is a slight splitting of the two subbands and the difference in their population becomes non zero. At the same time, it should be noticed that the position of both the subbands shifts to lower energy, so that, on the whole, the energy of the system is lowered by the splitting. For a small increase in $G$, the splitting increases and the increase in strain is very large. Further, the shifting of the subbands to lower energy is also large so that the energy of the system is much lower. Any further increase in $G$ does not have much effect on the quasiparticle spectrum any more which means the strain saturates. It should be emphasized that the strain is not introduced by hand but comes selfconsistently out of the model.

In Fig.5 we now fix the value of $G$ at 0.42, so that the model prefers the J-T split situation and look at the influence of $\langle S^z \rangle$. At saturation ($\langle S^z \rangle = S$), the spectral weight of $\uparrow$-subband is such that the $\alpha = 1$ subband is slightly more populated compared to the $\alpha = 2$ subband. As a result, a small strain appears. As $\langle S^z \rangle$ decreases, this spectral weight is modified in such a way that the occupation of the $\alpha = 1$ subband is more than that of the $\alpha = 2$. This results in an increase of the strain with $T$ as depicted in Fig. 2. This trend continues until $\langle S^z \rangle = 0$ and at that point, the strain is as if there is no exchange interaction in the model. In short, the effect of magnetization is to hinder J-T splitting.

In order to examine the interplay of magnetization and the strain as a function of $T$, it is necessary to include the temperature variation of both the strain and the magnetization. Therefore, we consider a specific case of $T_C = 250K$ and $T_s = 800K$ and the corresponding QDOS at different temperature are displayed in Fig. 6. Starting from $T = 0$, the $\alpha = 1$ subband occupation increases as $T$ increases up to $T_C$. Above $T_C$, the situation is reversed and both the $\alpha$-subbands are equally occupied at $T_s$. Therefore, the maximum of strain occurs at $T_C$ as shown in curve (b) of Fig. 2.

**IV. CONCLUSIONS**

The model Hamiltonian is solved by first absorbing the J-T term into the band electron term and then utilizing an interpolation ansatz for the selfenergy. The band splitting and through it the strain in the lattice due to J-T interaction has been determined selfconsistently. The strain as a function of the coupling constant at $T = 0$ is studied with and without the presence of local moment ordering. It is found that a minimum value of $G$ is required for the strain to appear. This critical $G$ is larger if the local moments are ordered or alternately if there is an external magnetic field. The temperature dependence of the strain is studied by assuming different values for the magnetic transition temperature. The study indicates that there is a strong interplay between the magnetic and structural transitions. It is observed that the

**FIG. 5:** Quasiparticle density of states(for lower sub-band in the positive half of the frame and for upper sub-band in the negative half of the frame) as a function of energy for various values of localized magnetization $\langle S^z \rangle$. Full line for spin up and dotted line for spin down. Thin vertical line shows the position of the chemical potential. $n = 0.7, J = 1, S = 3/2, C_0 = 1, T = 0$ and $G = 0.42$.

**FIG. 6:** Quasiparticle density of states(for lower sub-band in the positive half of the frame and for upper sub-band in the negative half of the frame) as a function of energy for various values of temperature $T$. Full line for spin up and dotted line for spin down. Thin vertical line shows the position of the chemical potential. $n = 0.7, J = 1, S = 3/2, C_0 = 1, T = 0$ and $G = 0.42$. 
growth of the strain appearing at $T_s (> T_C)$ is arrested with the onset of ferromagnetism and tends to a lower value determined by the magnetization at $T = 0$. This means the removal of spin degeneracy is not conducive to the removal of orbital degeneracy. The results are explained on the basis of the QDOS. The basic ingredients of the model are the band J-T effect, long range magnetic order and their mutual interaction. The possibility of different hopping between the degenerate bands is not considered for simplicity and therefore the results correspond to large J-T effect. The inter-orbital hopping, which is sometimes considered, would remove partially the degeneracy of the state. Also the magnetism which should evolve within the model is treated as a parameter. The detailed comparison with experiment is therefore not attempted. However, the general trends of the results related to suppression of J-T strain and coexistence of two phases are in tune with the experimental\textsuperscript{12,13,14} observations in a Ca-doped manganite and Heusler alloy. The calculation is based on the ansatz used for self-energy which is valid only in the limit of low charge carrier concentration. If this needs to be relaxed, the interaction among the band electrons has to be taken into account and naturally the ansatz for the selfenergy has to be modified.

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