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Theory of spin-phonon interaction effect on the Raman active peaks in manganite system

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Abstract. We report here a microscopic theoretical model showing the influence of spin-phonon interaction on Raman active peaks in the CMR manganite systems. This model Hamiltonian for the system consists of J-T distortion in e_g band, the double-exchange interaction and the Heisenberg spin-spin interaction among the core electrons. Further the phonons are coupled to e_g band electrons, J-T distorted e_g band as well as the double exchange interaction. The phonon Green's function is calculated by Zubarev's double time Green's function technique. The Raman spectral intensity is calculated from the imaginary part of the phonon Green's function. The intensity exhibits three Raman active peaks. The influence of spin-phonon coupling on these peaks will be discussed.

Keywords. Colossal magnetoresistance, Jahn-Teller distortion, Spin-phonon interaction

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1. Introduction

The physics of CMR is closely related to the features which are observed in the conducting transition-metal oxides with strong electron-electron and/or electron-lattice interactions. The correlated electrons, which are almost localized on the respective atomic sites attribute towards charge, spin and orbital degrees of freedom [1]. Further variety of phases like paramagnetic insulator, paramagnetic metal, ferromagnetic insulator and ferromagnetic metals are formed due to the variation of impurity concentration and temperature [2]. For the understanding of their electronic properties several mechanisms are proposed i.e double exchange

mechanism [3] Jahn-Teller (J-T) effect [4] and bipolaron formation [5]. Recently Rout and Parhi [6] reported that static J-T distortion produces a metal –insulator transition near the Curie temperature. Millis has suggested that the transition between the split e_g levels due to J-T distortion can be Raman active.

The experimental Raman studies display the phonon excitation [7 – 11] near the metal-insulator transition temperature. Recently Rout et.al. [12] have reported the theoretical study of Raman active CDW gap mode in manganites displaying the magnetic field and temperature dependence of the charge ordering peak. We propose here a model consisting of the double exchange model and the Heisenberg spin exchange interaction of the core electrons with J-T distortion as an extra mechanism. In order to study the evolution of the Raman active modes, the model Hamiltonian and the electron-phonon interaction are described in section 2. The calculation of the phonon Green's function and the Raman spectral intensity is given in section 3. The results and discussion are presented in section 4 and finally the conclusion is given in section 5.

2 The Model Hamiltonian

In doped manganites the three core t_{2g} electron spins are ferromagnetically coupled to the spin of the itinerant e_g electrons due to Hund's rule coupling. As an extra mechanism the orbital ordering is described by J-T effect which splits the degenerate e_g orbitals of Mn^{3+} ions. Further Heisenberg type direct spin-spin interaction is considered for the spins of the core electrons. The electronic part of the Hamiltonian is given as follows.

$$H_0 = \sum_{\alpha,k,\sigma} \epsilon_{k,\sigma} C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma} - \sum_{\alpha,k,\sigma} (-1)^{\alpha} Ge\, C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma} + \frac{1}{2} Ce^2 - J \sum_{\alpha,i} \overrightarrow{s_{\alpha,i}} \overrightarrow{S_i} - J_H \sum_{\langle i,j \rangle} \overrightarrow{S_i}.\overrightarrow{S_j} + \sum_{k,\sigma} (\epsilon_d - \sigma B^d) d^{\dagger}_{k,\sigma} d_{k,\sigma} \quad (1)$$

Within the mean field approximation the above Hamiltonian takes the form

$$H_0 = \sum_{\alpha,k,\sigma} \epsilon_{\alpha,k,\sigma} C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma} - \frac{1}{2} J\eta \sum_{\alpha,k,\sigma} \langle S^{\pm} \rangle\, C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\bar{\sigma}} - \frac{1}{2} J_H \sum_{k,\sigma} \langle S^{\pm} \rangle\, d^{\dagger}_{k,\sigma} + \sum_{k,\sigma} \epsilon_{d,\sigma}\, d^{\dagger}_{k,\sigma} d_{k,\sigma} \quad (2)$$

The first term describes the effective e_g electron interaction Hamiltonian with band energy

$$\epsilon_{\alpha,k,\sigma} = [\epsilon_0(k) - \mu - (-1)^{\alpha} Ge + M_1 \sigma] \quad (3)$$

Where $M_1 = \frac{1}{2} J M^d - B$ and $\epsilon_0(k) = -2t_0(cosk_x + cosk_y)$ with $2t_0$ is the nearest neighbor hopping integral and μ is the chemical potential. Due to J-T effect the e_g electron band splits with J-T gap energy of $2Ge$ where G and e are

respectively the static J-T coupling and lattice strain. Here B stands for external magnetic field with $\sigma = +1(-1)$ corresponding to spin up(down). The operator $C^{\dagger}_{\alpha,k,\sigma}(C_{\alpha,k,\sigma})$ is the creation (annihilation) operator of the conduction electron for the orbital $\alpha(= 1,2)$ designated for two split orbitals. Further J and M^d are respectively the exchange coupling and core magnetization respectively. The second term represents the spin fluctuation in e_g band with $\langle S^{\pm}\rangle$ representing the spin fluctuation parameter of the core electrons. Here η stands for a scale factor for the transverse exchange coupling. The third term represents the spin fluctuation interaction present in the core electrons with J_H as Heisenberg coupling. The last term represents the kinetic energy part of the core electron interaction with effective core electron level $\epsilon_{d,\sigma} = (\epsilon_d - \mu - \sigma + M_2^d\sigma)$, where $M_2 = \frac{1}{2}J_H M^d - B$ and the position of the core level is ϵ_d with respect to Fermi level $(\epsilon_F = 0)$. Here $d^{\dagger}_{k,\sigma}(d_{k,\sigma})$ represents the core electron creation (annihilation) operator.

In order to study the phonon properties, we consider here the phonon coupling to the band electrons as follows. The first term describes the phonon coupling to the degenerate conduction band electrons for the orbitals 1 and 2 with a normal electron phonon coupling f_1. The second term is the phonon coupling to the J-T split nondegenerate two orbital bands with a dynamic J-T coupling parameter f_2. Finally the third term is the phonon coupling to the double exchange interaction representing the spin- phonon interaction Hamiltonian with a spin phonon coupling f_3. Within mean-field approximation the interaction gives a term describing phonon coupling to electron densities of the e_g band electrons and the other phonon coupling to spin fluctuations in the conduction band. Finally the mean-field electron-phonon (EP) Hamiltonian appears as

$$H_{e-p} = \sum_{\alpha,k,\sigma,q}\lambda_{\alpha,\sigma}C^{\dagger}_{\alpha,k+q,\sigma}\,C_{\alpha,k,\sigma}A_q - \sum_{\alpha,k,\sigma,q}\eta f_3\,\langle S^{\pm}\rangle C^{\dagger}_{\alpha,k+q,\sigma}C_{\alpha,k+q,\bar{\sigma}}A_q \quad (4)$$

The effective EP coupling can be written as

$$\lambda_{\alpha,\sigma} = \left(f_1 - (-1)^{\alpha}f_2 e + f_3 M^d\sigma\right) \quad (5)$$

And the phonon displacement is given by $A_q = (b_q + b^{\dagger}_{-q})$ where $b^{\dagger}_q(b_q)$ is the phonon creation (annihilation) operator of the phonon wave vector q. Here the η is a scale factor multiplied to f_3 to represent phonon coupling to transverse spin fluctuation. Finally the bare-phonon Hamiltonian within mean-field approximation is written as

$$H_{e-p} = \sum_q \omega_q \, b_q^{\dagger} b_q \tag{6}$$

Where ω_q is the phonon frequency with phonon wave vector q.

3. Raman spectral density function

In order to explain Raman active peaks in the system in the Raman spectral intensity function, we calculate double time phonon Green's function defined as

$$D_{q,q'}(t,t') = i\theta(t,t') < \left[A_q(t); A_{q'}(t')\right] > \tag{7}$$

The phonon Green's function is evaluated from total Hamiltonian given in equation (2), (4) and (6). By applying Dyson approximation the Fourier transformed phonon Green's function is evaluated in the closed form as

$$D_{qq}(\omega) = \frac{\omega_q}{\pi[\omega^2 - {\omega_q}^2 - \sum(q,\omega)]} \tag{8}$$

The phonon self-energy is given by

$$\sum(q,\omega) = 4\pi\omega_q \, \chi_{qq}(\omega) \tag{9}$$

Where electron response function is written as

$$\chi_{q,q}(\omega) = \chi_1 + \chi_2 + \chi_3 + \chi_4 \tag{10}$$

Here $\chi_i (i = 1-4)$ are the two particle conduction electron Green's functions which are evaluated by taking only the mean-field electronic Hamiltonian H_0 given in eqn. (2) for simplicity of calculation. The spectral density function $I(\omega)$ is proportional to Raman spectral intensity and is calculated from the imaginary part of the phonon Green's function.

$$I(\omega) = -2\pi Im D_{q,q}(\omega + i\Gamma) \tag{11}$$

Where Γ represents the spectral width.Taking the real and imaginary part of the the self-energy with $(\omega \to \omega + i\Gamma)$, the spectral density function can be written as

$$I(\omega) = \frac{\omega_q Q}{P^2 + Q^2} \tag{12}$$

Where $P = \omega^2 - {\omega_q}^2 - Re\sum(q,\omega)$ and $Q = 2\Gamma\omega - Im\sum(q,\omega)$. The expression for P and Q are calculated explicitly for numerical computation.

The dimensionless EP couplings are

$$a_1 = \frac{N(0){f_1}^2}{\omega_0}, a_2 = \frac{N(0){f_2}^2}{\omega_0}, a_3 = \frac{N(0){f_3}^2}{\omega_0} \tag{13}$$

The physical parameters involved in the calculation are scaled by conduction band width (W). They are exchange coupling $g_1 = \frac{J}{W}$, J-T coupling $g = \frac{G}{W}$, temperature $t = \frac{k_{\beta,T}}{W}$, magnetic field $b = \frac{B}{W}$, frequency of ultrasound wave $c = \frac{\omega}{W}$, spectral width $ee = \frac{\Gamma}{W}$, and phonon – momentum transfer energy $q = \frac{qv_f}{W}$.

4. Results and Discussion

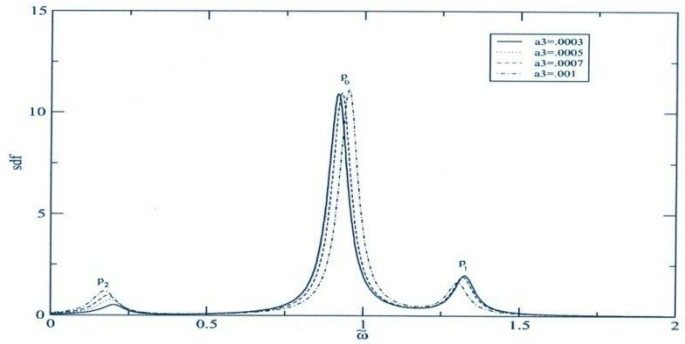

Fig. 1: The effect of spin-phonon coupling $a_3 = 0.0003\ to\ 0.001$ on the Raman active phonon peaks for fixed values of static J-T coupling $g = 0.0772$, double exchange coupling $g_1 = 0.01$, lattice strain $e = 0.802$, magnetization $m^d = 2.46$, conduction electron spin fluctuation $s = 0.061$, temperature $t = 0.02$, phonon momentum transfer energy $q = 0.03$, normal EP coupling $a_1 = 0.001$, dynamic J-T coupling $a_2 = 0.00001$

Fig.1 shows the effect of phonon coupling to the spin through the double exchange coupling for different values of spin-phonon coupling $\boldsymbol{a_3 = 0.0003}$ to 0.001. With the increase of spin-phonon coupling $\boldsymbol{a_3}$, the bare-phonon peak shifts to higher energies with the slight increase in spectral intensity exhibiting hardening behavior of phonon frequency. This hardening of phonon frequency has it's bearing on the magnetic excitation peak $\boldsymbol{p_1}$. With increase of $\boldsymbol{a_3}$, the position of the peak $\boldsymbol{p_1}$ shifts to lower energies with decrease in spectral intensity showing the decrease of the magnetization with increase of spin-phonon coupling. On the other hand, the phonon-momentum transfer energy shifts to the lower energies with increase in spectral intensity .The evolution of the peaks $\boldsymbol{p_0}$, $\boldsymbol{p_1}$ and $\boldsymbol{p_2}$ display a strong interplay between the lattice energy and magnetic energy through the spin-phonon coupling $\boldsymbol{a_3}$. Dediu et. al. [13] have reported the strong charge and spin-lattice coupling present in their study for $\boldsymbol{Pr_{0.65}Ca_{0.35}MnO_3}$ in Raman spectra. Similarly Gupta et. al. [14,15] have reported the evolution of Raman active $\boldsymbol{A_g}$ phonon mode in

$Pr_{0.63}Ca_{0.37}MnO_3$as a function of temperature, revealing that their frequencies and unexpectedly line widths increase as temperature decreases due to strong spin-phonon coupling. The details of the other phenomenon will be published elsewhere.

5. Conclusions

We have proposed a model Hamiltonian for the manganites which consists of the J-T distortion e_g band, the double exchange between itinerant core electrons and the localized t_{2g} electrons in presence of a Heisnberg spin-spin interaction between the core electrons. For a suitable set of model parameters the Raman spectra displays three peaks i.e bare-phonon peak p_0, magnetic excitation peak p_1, and phonon momentum transfer excitation peak p_2.The effect of spin-phonon coupling on the Raman active peaks are discussed.

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