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Raman active peaks in heavy fermion systems: a model study of the Coulomb interaction effect in ferromagnetic limit

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Abstract. We report here a model study of Raman spectra in heavy fermion systems in normal state. We consider Periodic Anderson Model along with phonon coupling to hybridization between the conduction and f-electrons as well as phonon coupling to the bare f-electrons and phonon vibrational energy within harmonic approximation. The Coulomb interaction between f-electrons in the ferromagnetic limit, is considered within the Hartee-Fock type mean-field approximation. The phonon Green's function is calculated by using Zubarev's Green's function technique. Phonon spectral density function (SDF) is calculated from the imaginary part of the phonon Green's function applying a small phonon spectral width. The intensity of Raman spectra is proportional to the phonon SDF. The Raman spectra display a bare phonon peak as well as a hybridization gap excitation peak. Evolution of these peaks are investigated by varying the model parameters of the system.

Keywords. Electron-phonon interaction, Raman spectra, Coulomb interaction, Ferromagnetism

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1. Introduction

Raman scattering is a useful technique to study ground state, low energy excited states as well as the lattice anomalies in heavy fermion systems (HFS). Raman scattering has been used to study crystalline electric-field excitations and localized excitation electronic excitations in $CeCu_2Si_2$and UPt_3[1-4]. Raman

scatteringby electrons can be used to get information on the structure of the Fermi surface, electron velocities, and electron scattering mechanisms[5, 6]. Also, It has been used to study valence fluctuations and heavy fermion behavior in metallic compounds of the rare earths and actinides [7, 8] and the information corresponding to $q \approx 0$ are complementary to neutron scattering. Raman scattering in UPt_3[9] at300 K, 77 K and 5K**,** due to spin fluctuations is of magnetic origin**.** The magnetic ordering temperature in some Cerium based heavy fermion compounds lies within a low temperature range i.e.5K-10K, while ferromagnetic order is present in uranium based heavy fermion compounds such as UTe, $UCuSb_2$at Curie temperature T_C is as high as 102K or 113K. In neptunium compounds, $NpNiSi_2$and Np_2PdGa_3 with $T_C$equal to 51.5K and 62.5K,respectively [10]. Due to Coulomb repulsive interaction, in HFS, the f-electrons are localized, and exhibit localized magnetic moments. The interaction between these localized magnetic moments and conduction electrons gives rise to long range magnetic orders. Raman scattering from phonons can be used to study electron-phonon (EP) interaction and to get essential clues for understanding of the mechanism of superconductivity in HFS.

Rout et al. have reported the theoretical studies of the Raman spectra in normal state as well as in superconducting phase of HFS [11,12]. More recently, Rout et al. [13,14] have studied the effect of paramagnetic limit of Coulomb interaction on the temperature anomaly in velocity of sound and ultrasonic attenuation in HFS. In present model study of Raman spectra in heavy fermion systems, in normal state, we have considered Periodic Anderson Model along with phonon coupling to hybridization between the conduction and f-electrons as well as phonon coupling to the bare f- electrons and phonon vibrational energy within harmonic approximation. The Coulomb interaction between f-electrons in the ferromagnetic limit, is considered within the Hartee-Fock type mean-field approximation.

2. Formalism

The heavy fermion system is described by the Periodic Anderson Model by the Hamiltonian which is written as

$$H_1 = \sum_{k,\sigma}(\epsilon_k - \mu - \sigma B_c)\, c^{\dagger}_{k\sigma} c_{k\sigma} + \sum_{k,\sigma}(\varepsilon_f - \sigma B_f) f^{\dagger}_{k,\sigma} f_{k,\sigma} + V \sum_{k,\sigma}\left(f^{\dagger}_{k,\sigma} c_{k,\sigma} + c^{\dagger}_{k,\sigma} f_{k,\sigma}\right) + U \sum_{k,\sigma} n^{f}_{i\uparrow} n^{f}_{i\downarrow} \quad (1)$$

Here the first term describes kinetic energy interaction of conduction electron where$\epsilon_k\mu$, and B_c are respectively the conduction electron energy, chemical potential and the magnetic field applied to conduction electrons. The second term describes the f-electron kinetic energy term with $\epsilon_f$as the f-electron energy and B_f as magnetic field applied to f-electrons. The third term describes the hybridization between conduction and f-electrons with hybridization strength V. Further, the last term represents the electron correlation between localized f-electron with U as the Coulomb energy. The Coulomb correlation is treated herewithin Hartree-Fock mean-field approximation in ferromagnetic limit and can be written as $Un_{i\uparrow}^{f}n_{i\downarrow}^{f} \rightarrow U < n_{i\uparrow}^{f} > n_{i\downarrow}^{f} + U < n_{i\downarrow}^{f} > n_{i\uparrow}^{f} - U < n_{i\uparrow}^{f} >< n_{i\downarrow}^{f} >$.Taking Fourier transformation, the Coulomb term can be written as $U\sum_{k,\sigma} < n_{-\sigma}^{f} > n_{k\sigma}^{f}$ with the average f-electron occupation number written as $< n_{-\sigma}^{f} > = \frac{1}{N}\sum_{k,\sigma} < f_{k,-\sigma}^{\dagger} f_{k,-\sigma} >$. With the approximation, the Hamiltonian in eq.(1) is written as

$$H_1 = \sum_{k,\sigma}\epsilon_{k\sigma} c^{\dagger}_{k,\sigma} c_{k,\sigma} + V\sum_{k,\sigma}\left(f^{\dagger}_{k,\sigma} c_{k,\sigma} + c^{\dagger}_{k,\sigma} f_{k,\sigma}\right) + \sum_{k,\sigma} E_{0\sigma} f^{\dagger}_{k,\sigma} f_{k,\sigma}. \quad (2)$$

where$\epsilon_{k\sigma} = (\epsilon_k - \mu - \sigma B_c)$ and $E_{0\sigma} = (\varepsilon_f - \sigma B_f + U < n_{-\sigma}^{f} >)$.The phonon coupling to hybridization between c-and f-electrons in the presence of phonon vibration with phonon frequency ω_q is written as

$$H_2 = \sum_{k,q,\sigma}[f_1(q)(c^{\dagger}_{k+q,\sigma} f_{k,\sigma} + f^{\dagger}_{k+q,\sigma} c_{k,\sigma}) + f_2(q)(f^{\dagger}_{k,\sigma} f_{k,\sigma})]A_q + \sum_q \omega_q b_q^{\dagger} b_q \quad (3)$$

where$f_1(q)$ and $f_2(q)$are the EP couplings to hybridization and conduction electrons respectively. $A_q = b_q + b_{-q}^{\dagger}$is the phonon displacement with $b_q(b^{\dagger})$ as the phonon annihilation (creation) operator. The Fourier transformed phonon Green's function is calculated by Zubarev'sGreen's function technique [15] and is written as

$$D_{q,q}(\omega) = \ll A\_q\,(t); A\_q\,(t') \gg_{\omega} = (\omega_q/\pi)[\omega^2 - \omega_q^2 - \sum(\omega, q)]^{-1} \quad (4)$$

where the phonon self-energy, $\sum(\omega, q) = 4\pi\omega_q\chi_{q,q}(\omega)$ with electron response function$\chi_{q,q}(\omega)$, which is calculated from the Hamiltonian H_1 only. The electron response function $\chi_{q,q}(\omega)$ is a two particle Green's function. The calculation of $\chi_{q,q}(\omega)$ involves higher order two particle Green's functions. The coupled Green's function are solved in the closed form in integral form. The spectral

density function (SDF) which is proportional to Raman scattering intensity, is calculated for long wave length limit (q→0) of phonons at a finite temperature by attributing a finite width (η) to phonon frequency i.e. SDF $= -2\,\mathrm{Im}\,\mathrm{D}_{\mathrm{qq}}(\omega + i\eta)_{\mathrm{q}=0}$. The SDF is computed numerically by varying the physical parameters of the HFS. All the physical parameters are made dimensionless with respect to Debye energy ω_D.The dimension less parameters are EP coupling to hybridization $\mathrm{g}=\{\mathrm{N}(0)\mathrm{f}_1^2(\mathrm{o})\}/(\omega_0)$, the phonon coupling to the f-electron $\mathrm{g_f} = N(0)f_2^2/\omega_\mathrm{D}$ with $r = \sqrt{\mathrm{g_f}/\mathrm{g}}$, position of f-level $\mathrm{d} = \varepsilon_\mathrm{f}/\omega_\mathrm{D}$, Coulomb interaction energy $\mathrm{u} = \mathrm{U}/\omega_\mathrm{D}$, strength of hybridization $\mathrm{v} = \mathrm{V}/\omega_\mathrm{D}$, temperature$\mathrm{t} = \mathrm{k_B T}/\omega_\mathrm{D}$, phonon frequency $\mathrm{p} = \omega_{0/}\omega_\mathrm{D}$, spectral width $\mathrm{e} = \eta/\omega_\mathrm{D}$ and renormalized frequency, $c = \omega/\omega_\mathrm{D}$ with reduced frequency $\widetilde{\omega} = \omega/\omega_0$.

3. Results and Discussion

The f- electron occupancies i.e. $< \boldsymbol{n}_{\uparrow}^{\boldsymbol{f}} >$ for spin up electrons and $< \boldsymbol{n}_{\downarrow}^{\boldsymbol{f}} >$ for spin down electrons are calculated from the mean-field electron Hamiltonian $\boldsymbol{H_1}$ given in equation(2) and are solved numerically and self-consistently for different values of position of f-electron (d) and Coulomb energy (u). The plots are shown in fig.1.

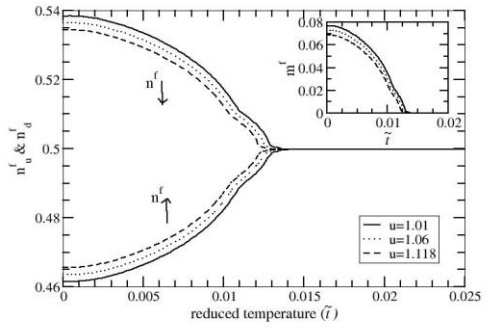

Fig.1. The plots of $\boldsymbol{n}_{\uparrow}^{\boldsymbol{f}}$ and $\boldsymbol{n}_{\downarrow}^{\boldsymbol{f}}$ (determined self- consistently) vs. reduced temperature $(\tilde{\boldsymbol{t}})$ and in the inset, plots of magnetization $\boldsymbol{m^f}$ vs. reduced temperature $(\tilde{\boldsymbol{t}})$ for different values of Coulomb correlation energy $\boldsymbol{u} = \mathbf{1.01}, \mathbf{1.06}, \mathbf{1.18}$ in the ferromagnetic limit.

This shows that the temperature dependent f-electron occupancies are in-equivalent i.e.$\boldsymbol{n}_{\downarrow}^{\boldsymbol{f}} > \boldsymbol{n}_{\uparrow}^{\boldsymbol{f}}$, leading to ferromagnetic ordering in the system up to the

Curie temperature $\boldsymbol{t_c}$.The occupancies are same i.e. $< \boldsymbol{n_\uparrow^f} > = < \boldsymbol{n_\downarrow^f} > = \mathbf{1/2}$ in paramagnetic phasefor temperature$> \boldsymbol{t_c}$. The temperature dependent magnetization is shown in the inset of fig.1.We have taken the occupancies $< \boldsymbol{n_\uparrow^f} >$= 0.46424 and $< \boldsymbol{n_\downarrow^f} >$= 0.53636 atreduced temperature $\boldsymbol{t = 0.0025}$ for reduced Coulomb energy $\boldsymbol{u = 1.06}$ from thefig.1. Using these values we have computed the spectral density function (SDF) and SDF isshown in fig. 2 (a) for EP coupling to hybridization, $\mathbf{g = 0.00}$, $\mathbf{g = 0.011}$.In absence of EP coupling, $\mathbf{g = 0.00}$, phonon self-energy becomes zero and hence, we observe a single peak ($\boldsymbol{P_0}$) at energy $\boldsymbol{\tilde{\omega} = \omega/\omega_0 = 1}$, corresponding to the bare phonon vibrational frequency $\boldsymbol{\omega_0}$ of the ferromagnetically ordered HFS.

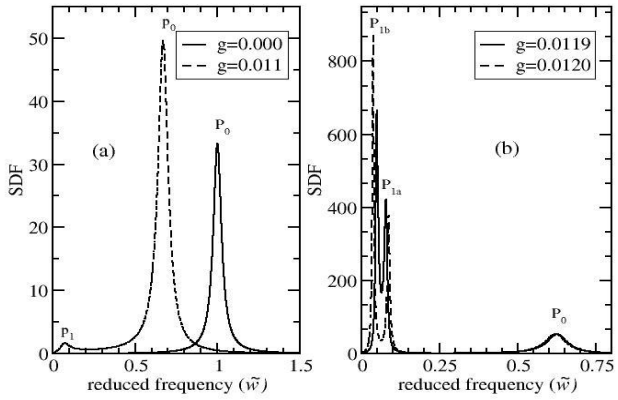

Fig. 2. The plotsof spectral density function (SDF) vs. reduced frequency ($\tilde{\boldsymbol{\omega}}$) for different values of electron-phonon coupling to hybridization in fig. (a) g = 0.0 and 0.011 in fig. 1(b), g = 0.0119 and g = 0.012 with fixed values of r = 0.001,d =-u/2,v = 0.03, u = 1.06, t = 0.0025, e = 0.03,b=0.0 and p =1.0.$\boldsymbol{n_\uparrow^f}$= 0.46424 and $\boldsymbol{n_\downarrow^f}$= 0.53636.

On increase the EP coupling $\mathrm{g} = 0.011$, it is expected that the phonons acquire self-energy due to the interaction with electrons in the system. For a minimum value of EP coupling to $\mathrm{g} = 0.011$, we just observ a low intensity peak (P_1) at lower energy beside the bare phonon peak P_0 . Under this condition, the peak P_1 appears at energy $\tilde{\omega} = 0.077072$ associated with the hybridization between the conduction electrons and the localized f-electrons. It is to note further that, the bare phonon peak P_1 shifts from ω_0 = 1 to ω_0 = 0.670956 indicating the strong renormalization of the phonon frequency and exhibiting its

softening behavior. The present minimum EP coupling, g = 0.011 in the ferromagnetic phase is smaller than the EP coupling g $= 0.024$ in paramagnetic phase [16] and g = 0.0253 (in normal phase of HFS) [12].On further increasing the EP coupling to g $= 0.0119$and g $= 0.0120$, the hybridization peak (P_1) splits into two (peaks P_{1a} and P_{1b}). The high energy peak (P_{1a}) with lower spectral height and wider spectral width is associated with the conduction band while the low energy sharp peak (P_{1b}) with narrow spectral width is associated with the localized f-electrons. These two peaks are separated by the hybridization gap, which is modulated strongly with EP coupling. We conclude that the EP coupling strongly renormalizes the hybridization gap in HFS.

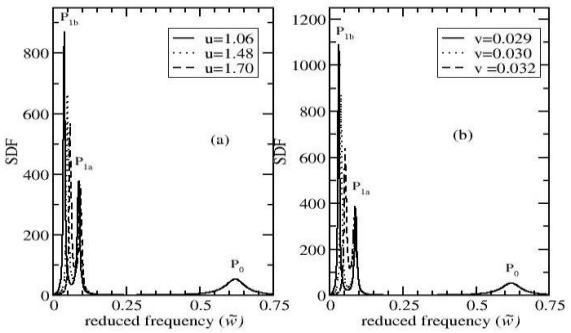

Fig. 3. The plots of spectral density function (SDF) vs. reduced frequency($\tilde{\omega}$) in (a) for different values of Coulomb interaction energy u =1.06, 1.48, and 1.70 with fixed values of g = 0.012, r=0.001, v=0.03, d=-u/2, t=0.0025, e = 0.03, b =0.0 and p =1.0, $n_{\uparrow}^{f}$= 0.46424 and $n_{\downarrow}^{f}$= 0.53636, in (b) for different values of strength of hybridization v = 0.029, 0.30 and 0.032 with fixed values of g = 0.012, r=0.001, u = 1.06, d=-u/2, t=0.002, e = 0.03 ,b = 0.0 and p =1.0.$n_{\uparrow}^{f}$= 0.46424 and $n_{\downarrow}^{f}$= 0.53636.

The Coulomb interaction between f-electrons, renormalizes the hybridization gap as well as the position of f-level with respect to Fermi level in the heavy fermion systems. Within Hartee-Fock mean-field approximation,in the ferromagnetic limit, the effective f-level position for spin σ appears as $E_{0\sigma} = \varepsilon_f + U < n_{-\sigma}^{f} >$ which appears as $d_1 = d + u * (< n_{\downarrow}^{f} > - < n_{\uparrow}^{f} >)$ where d is the bare f-electron position. At a given temperature, the mean f-electron occupancies i.e. $< n_{\uparrow}^{f} >$ and $< n_{\downarrow}^{f} >$are taken from the temperature dependent $< n_{\uparrow}^{f} >$ and $< n_{\downarrow}^{f} >$ which are computed self-consistently. Under half filled condition for the f-level energy ,i.e. $d = -u/2$, the effect of Coulomb

interaction on the Raman active peaks for different values of $u = 1.06 - 1.70$ is as shown in fig.3. For a given temperature t = 0.0025, and the ferromagnetic limit of Coulomb correlation energy $u = 1.06$,$< n_{\uparrow}^{f} > = 0.46242$ and $< n_{\downarrow}^{f} >$ = 0.53636 and bare f-electron position $d = -0.53$, the renormalized f-electron position becomes d_1 = 0.53+(0.46242 −0.53636)1.06 =- 0.6083764. Thus, within the ferromagnetic limit of Coulomb interaction, the renormalized f-electron position is enhanced linearly with u. As a result, the hybridization peaks (P_{1a}, P_{1b}) gradually approach each other more and more with increase of Coulomb interaction. The peak P_{1a}does not change its position with increase in intensity, while the peak P_{1b} shifts to higher energies with decrease in intensity. On the other hand, the phonon peak (P_0) is not affected with increase of Coulomb energy. Fig. 3(b) shows the effect of strength of hybridization on Raman active peak. With the increase of strength of hybridization (v) the hybridization gap decreases with the decrease in the peak intensities.

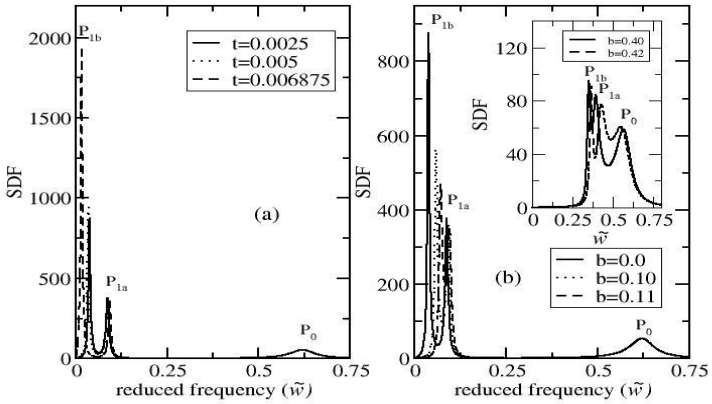

Fig. 4. The plots of spectral density function (SDF) vs. reduced frequency ($\tilde{\omega}$) in (a) for different values of temperature t = 0.0025, 0.005 and 0.006875 with($n_{\uparrow}^{f}$= 0.46424 and $n_{\downarrow}^{f}$= 0.53636), ($n_{\uparrow}^{f}$= 0.46848 and $n_{\downarrow}^{f}$= 0.53212) and($n_{\uparrow}^{f}$= 0.4709 and $n_{\downarrow}^{f}$= 0.5209) at this temperatures respectively with fixed values of u =1.06, d =-u/2, g = 0.012, r = 0.001, v = 0.03, , e = 0.03, b = 0.0 and p = 1.0. In fig.(b) for different values of magnetic field b = 0.0, 0.10, 0.11 and in the inset for b=0.40, 0.42 with fixed values g = 0.012, r = 0.001, v = 0.03, , e = 0.03, and p =1.0, $n_{\uparrow}^{f}$= 0.46424 and $n_{\downarrow}^{f}$= 0.53636

The effect of temperature on Raman spectra is shown in fig. 4(a). The difference between f-electron occupancies i.e. ($< \boldsymbol{n}^{\boldsymbol{f}}_{\downarrow} >- < \boldsymbol{n}^{\boldsymbol{f}}_{\uparrow} >$) is small near Curie temperature i.e. $\boldsymbol{t_c}$= 0.01375 leading to weak magnetization. Hence, the SDF shows the hybridization gap between the peaks $\boldsymbol{P_{1a}}$ and $\boldsymbol{P_{1b}}$of magnitude 0.0495704at t=0.0025. The difference between f-electron occupancies is very large at temperature t=0 leading to very strong ferromagnetic order. Hence, we observe that the SDF shows a small separation between the hybridization peaks$\boldsymbol{P_{1a}}$ and $\boldsymbol{P_{1b}}$ at t=0. Hence, we conclude that at lower temperatures the hybridization gap is suppressed due to ferromagnetic order in the HF systems.However, at low temperature, below Curie temperature, the bare phonon peak $\boldsymbol{P_0}$ is not effected by the temperature.

Fig. 4(b) shows the effect magnetic field on the Raman active peaks in ferromagnetically ordered heavy fermion systems. In absence of external magnetic field, b = 0.0, the peaks $\boldsymbol{P_{1a}}$ and $\boldsymbol{P_{1b}}$ are separated by the hybridization gap of the systems. The sharp peak $\boldsymbol{P_{1b}}$ associated with the band having localized f-electron character and the relatively wide peak $\boldsymbol{P_{1a}}$exhibits the conduction electron character. On the application of external magnetic field both peaks shift to higher energies with reduced spectral height of both the peaks. The hybridization gap is also reduced with the application of the magnetic field for $\boldsymbol{b = 0.02}$to $\boldsymbol{0.11}$. For very high magnetic fields i.e. $\boldsymbol{b = 0.40, 0.42}$, the hybridization peaks $\boldsymbol{P_{1a}}$ and $\boldsymbol{P_{1b}}$ move towards the phonon peak and they are very strongly coupled to phonon vibrations. Hence their peak heights are suppressed considerably(nearly 10 times).

4. Conclusion

We have considered Periodic Anderson Model in presence of EP interaction. We have calculated Raman spectral intensity from the imaginary part of phonon Green's function. In this spectra, we observe a phonon peak and a hybridization gap peak which splits into two. We investigate the effect of EP coupling, Coulomb interaction, hybridization strength and temperature and magnetic field on the Raman active peaks.

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