Phonon frequency shift and effect of correlation on the electron–phonon interaction in heavy fermion systems

B OJHA, P NAYAK and S N BEHERA*

P.G. Department of Physics, Sambalpur University, Jyoti Vihar, Burla 768 019, India
*Institute of Physics, Sachivalaya Marg, Bhubaneswar 751 005, India

Abstract. The electron–phonon interaction in the periodic Anderson model (PAM) is considered. The PAM incorporates the effect of onsite Coulomb interaction (U) between f-electrons. The influence of Coulomb correlation U on the phonon response of the system is studied by evaluating the phonon spectral function for various parameters of the model. The numerical evaluation of the spectral function is carried out in the long wavelength limit at finite temperatures keeping only linear terms in U. The observed behaviour is found to agree well with the general features obtained experimentally for some heavy fermion (HF) systems.

Keywords. Periodic Anderson model (PAM); heavy fermion system; electron–phonon interaction; Coulomb correlation; double time temperature dependent Green function.

PACS Nos 74.70.Tx; 71.10.-w; 71.38.+i

1. Introduction

The experimental investigations on some intermetallic compounds and alloys of rare earths like Ce, Yb and actinides like uranium have revealed very interesting physical properties which can be associated with their partially filled shell of f-electrons. These systems are known to form highly correlated electronic states which exhibit unusual magnetic, thermo-dynamic, transport properties in their normal state besides exhibiting superconductivity at low temperatures. Heavy fermions belong to one such group and have attracted considerable interest in recent years [1–4]. It is also established that all the anomalous properties exhibited by the heavy fermions can be understood as arising from the strong hybridization of the correlated f-electrons with those in the conduction band near the Fermi level.

The results of some recent experiments ([5] and references therein) suggest that in these HF systems there exist a strong coupling of the elastic degrees of freedom with those of the electronic and magnetic ones. Particularly the physical properties related to the observed magneto-elastic effect (coupling of phonon to the f-electrons), anisotropic Fermi surface, Kondo volume collapse etc and the manifestation of the deformation potential (the coupling of phonons to the conduction electrons), have persuaded many to emphasize the role of the electron–phonon interaction [2,5] to explain some of the low temperature

behaviour. Though direct experimental evidence for phonon anomaly through inelastic neutron scattering or Raman scattering experiments are rare in these systems, however, the measurements on elastic constant, ultrasonic attenuation and sound velocity have provided indirect evidence of strong electron–phonon coupling. The most prominent of these are the softening of the elastic constants, phonon frequencies and observation of anomalies in thermal expansion with temperature below the Kondo temperature. The bulk modulus and elastic constant measurements at low temperature have shown prominently the existence of strong phonon anomalies in $CeAl_3$, $CeCu_6$, UPt_3, UBe_{13}, $CeRu_2 Si_2$ and few others which give an indirect evidence of the electron–phonon coupling in these systems [5–7]. Some uranium based HF systems also show phonon anomalies due to anharmonic interaction with zone boundary phonons and exhibit soft mode behaviour [5].

Over the last thirty years the periodic Anderson model (PAM) [9] has emerged as one of the most successful models for explaining many of the properties of the correlated systems [8,1,3,10]. It is believed that, the intra-atomic Coulomb interaction between the f-electrons plays a dominant role for explaining the properties of the HF systems. The strong correlation between the f-electrons imposes rather strong local restrictions on the electronic occupancy to be either zero or one at each site. Taking into account the effect of finite band width, Lee *et al* [11] have established that the electron–phonon coupling constant becomes dominant for nearly filled band in the strongly correlated limit. The study of Miyake *et al* [12,13] and a more recent study of Min *et al* [14] have concluded through their calculations that a phonon mediated BCS type superconductivity is possible in HF systems. The later study have also revealed that the Coulomb correlation results in the hardening of the phonon and the enhancement of the strength of electron–phonon coupling constant. Considering the important role played by the electron correlation, in this paper we have analysed its effect on different phonon anomalies by calculating the phonon response function as a function of temperature.

In this paper, followign Fulde *et al* [1], we consider two different electron–phonon coupling mechanisms in the framework of the periodic Anderson model (PAM). These are (i) the usual interaction between the phonons with the electrons in the f-bands and (ii) the electron–phonon interaction arising from hybridization term of the (PAM). Our aim in this paper is to evaluate the contribution to the phonon self-energy from the mixing of the f and conduction electrons as well as the f electrons alone in the presence of the onsite Coulomb repulsion. For simplicity, we have first explored the properties of the system in the long wavelength limit, for small temperature retaining terms only linear in correlation.

The plan of the rest of the paper is as follows. The formalism and the calculations are given in §2. In §3 we discuss the results. Finally we conclude in §4.

2. Formalism

As mentioned in the introduction a description of the phonon anomalies in the HF system will require a total Hamiltonian

$$H = H_0 + H_{e-p} + H_p \tag{1}$$

which consists of three terms: (i) the electronic Hamiltonian H_0, (ii) the Hamiltonian for the phonons H_p and (iii) the electron–phonon interaction term H_{e-p}. The electronic Hamiltonian H_0 correspond to that of the PAM and is given by

$$H_0 = \sum_{k\sigma} \varepsilon_k C_{k\sigma}^+ C_{k\sigma} + E_0' \sum_{k\sigma} f_{k\sigma}^+ f_{k\sigma} + \gamma_0 \sum_{k\sigma} (f_{k\sigma}^+ C_{k\sigma} + C_{k\sigma}^+ f_{k\sigma})$$

$$+ (U/2) \sum_{i\sigma} n_{i\sigma}^f n_{i-\sigma}^f, \tag{2a}$$

where $C_{k\sigma}^+$ ($C_{k\sigma}$) and $f_{k\sigma}^+$ ($f_{k\sigma}$) are the creation and annihilation operators for conduction and f-electrons with momentum k and spin σ respectively, ε_k is the energy of electron in the conduction band, E_0' is the position of f-level, $n_\sigma = f_{i\sigma}^+ f_{i\sigma}$ is the number operator for f electrons, γ_0 represent the strength of the hybridization between the f-electrons and the conduction electrons and U is the onsite Coulomb repulsion between the f-electrons of opposite spin.

Considering the importance of the lanthanide contraction in these systems, the phonons are assumed to interact predominantly with the f-electrons. While the interaction of the phonons with the conduction electrons is neglected, it has been argued by Fulde that their interaction with the hybridization term can contribute substantially to the phonon anomalies. Thus the electron–phonon interaction Hamiltonian is given by

$$H_{e-p} = \sum_{kq\sigma} \Big[f_1(q)(f_{k+q,\sigma}^+ C_{k,\sigma} + C_{k+q,\sigma}^+ f_{k,\sigma})$$

$$+ f_2(q) f_{k+q,\sigma}^+ f_{k,\sigma} \Big] [b_q + b_{-q}^+], \tag{2b}$$

where $f_1(q)$ and $f_2(q)$ are the coupling constants, the former corresponds to the interaction arising between phonons with the hybridization terms and the latter corresponds to the strength of interaction with the f-electrons. Finally the Hamiltonian for the phonons is given by

$$H_p = \sum_q \omega_q b_q^+ b_q, \tag{2c}$$

b_q^+ (b_q) being the creation (annihilation) operator for the phonons with the wave vector q and frequency ω_q.

Since we are interested in the calculation of phonon response functions, it is necessary to evaluate the phonon Green function [15] defined as

$$D_{qq'}(t - t') = \langle\langle A_q(t); A_{q'}(t')\rangle\rangle = -iv(t - t')\langle[A_q(t); A_{q'}(t')]_-\rangle, \tag{3}$$

where

$$A_q = b_q + b_{-q}^+ \quad \text{and} \quad B_q = b_q - b_{-q}^+ \tag{4}$$

are respectively the qth Fourier component of the displacment and momentum of the ions.

The phonon Green function (eq. (3)) can be calculated by writing its equation of motion using the Hamiltonian of eq. (1), which when Fourier transformed $D_{qq'}(\omega)$, can be expressed in the form

$$D_{qq'}(\omega) = \delta_{-qq'}(\omega_q/\pi)[\omega^2 - \omega_q^2 - 4\pi\omega_q \chi_{qq}(\omega)]^{-1}, \tag{5}$$

where the Fourier transforms of the response functions entering in the self-energy is given by

$$\chi_{qq'}(\omega) = f_1(-q)f_1(-q')\Gamma_3(qq'\omega) + f_1(-q)f_2(-q')\Gamma_4(qq'\omega)$$
$$+ f_2(-q)f_1(-q')\Gamma_5(qq'\omega) + f_2(-q)f_2(-q')\Gamma_6(qq'\omega), \tag{6}$$

where Γ_i's ($i = 3$ to 6) represent the electron response functions. These electron response functions are higher order Green functions of the electron operators and are evaluated from the equations of motion of these Green functions using only the electronic Hamiltonian given by eq. (1) without the phonon and the electron–phonon interaction term i.e.

$$H_0 = \sum_{k\sigma} \varepsilon_k C^\dagger_{k,\sigma} C_{k,\sigma} + E'_0 \sum_{k\sigma} f^\dagger_{k,\sigma} f_{k,\sigma} + \sum_{k\sigma} \gamma_0 (f^\dagger_{k,\sigma} C_{k,\sigma} + C^\dagger_{k,\sigma} f_{k,\sigma})$$
$$+ (U/2) \sum_{i\sigma} n^f_{i\sigma} n^f_{i-\sigma}. \tag{7}$$

In the long wavelength limit i.e. for $q = 0$, the expressions for these response functions are given by

$$\Gamma_3(q = 0, \omega) = \sum_{k\sigma k'\sigma'} \Gamma_3(k, k', \omega)$$

$$= (\delta_{\sigma\sigma'}\delta_{kk'}/2\pi|D|) \Big[2(\omega - \varepsilon_k + E'_0)(\langle C^\dagger_{k,\sigma} C_{k,\sigma'} \rangle - \langle f^\dagger_{k,\sigma} f_{k,\sigma'} \rangle)$$

$$+ 2(U/2)\langle n^f_{k-\sigma} \rangle \langle C^\dagger_{k,\sigma'} C_{k,\sigma} \rangle - \langle f^\dagger_{k,\sigma'} f_{k,\sigma} \rangle) \Big], \tag{8a}$$

$$\Gamma_4(q = 0, \omega) = (\delta_{\sigma\sigma'}\delta_{kk'}/2\pi|D|) \Big[(\omega - \varepsilon_k + E'_0)\langle C^\dagger_{k,\sigma} f_{k,\sigma'} \rangle$$

$$- (\omega + \varepsilon_k - E'_0)\langle f^\dagger_{k,\sigma} C_{k,\sigma} \rangle \Big], \tag{8b}$$

$$\Gamma_5(q = 0, \omega) = (\delta_{\sigma\sigma'}\delta_{kk'}/2\pi\omega|D|)\gamma_0 \Big[2\omega(\langle f^\dagger_{k,\sigma} f_{k,\sigma'} \rangle - \langle C^\dagger_{k,\sigma} C_{k,\sigma'} \rangle) \Big], \tag{8c}$$

$$\Gamma_6(q = 0, \omega) = (\delta_{\sigma\sigma'}\delta_{kk'}/2\pi\omega|D|)\gamma_0 \Big[-(\omega - \varepsilon_k + E'_0)\langle C^\dagger_{k,\sigma} f_{k,\sigma'} \rangle$$

$$- (\omega + \varepsilon_k - E'_0)\langle f^\dagger_{k,\sigma} C_{k,\sigma'} \rangle \Big], \tag{8d}$$

where

$$|D| = (\omega + \varepsilon_k - E'_0)(\omega - \varepsilon_k + E'_0) - 4\gamma_0^2 - (U/2)\langle n^f_{k-\sigma} \rangle(\varepsilon_k - E'_0). \tag{9}$$

Following the standard technique of Zubarev [15], the different correlation functions involved in these equations are evaluated. The expressions of these are given by

$$\langle C^\dagger_{k\sigma} C_{k\sigma} \rangle = -y_{12} \Big[y_{11}(y_1 - \tilde{E}'_0) - y_{22}(y_2 - \tilde{E}'_0) \Big], \tag{10a}$$

$$\langle f^\dagger_{k\sigma} f_{k\sigma} \rangle = -y_{12} \Big[y_{11}(y_1 - \varepsilon_k) - y_{22}(y_2 - \varepsilon_k) \Big], \tag{10b}$$

$$\langle C^\dagger_{k\sigma} f_{k\sigma} \rangle = \langle f^\dagger_{k\sigma} C_{k\sigma} \rangle = y_{12}\gamma_0 \Big[y_{11} + y_{22} \Big], \tag{10c}$$

where

$$y_{11} = \frac{1}{e^{y_1/\theta} + 1}; \quad y_{22} = \frac{1}{e^{y_2/\theta} + 1}; \quad y_{12} = \frac{-\delta_{kk'}\delta_{\sigma\sigma'}}{(y_1 - y_2)}, \tag{11a}$$

$$y_1 = (1/2)(\varepsilon_k + \tilde{E}'_0) + (1/2)\sqrt{(\varepsilon_k - \tilde{E}'_0)^2 + 4\gamma_0^2}, \tag{11b}$$

$$y_2 = (1/2)(\varepsilon_k + \tilde{E}'_0) - (1/2)\sqrt{(\varepsilon_k - \tilde{E}'_0)^2 + 4\gamma_0^2} \tag{11c}$$

and

$$\tilde{E}'_0 = E'_0 + (U/2)\langle n^f_{k,-\sigma}\rangle. \tag{12}$$

The values of the time correlation function are substituted in eq. (9) to get the final form of the different Green functions which involve different parameters of the system. These evaluated quantities are further resubstituted back in eqs (6) and (5) to get the expression for the phonon Green function $D_{qq'}(\omega)$ sought for the calculation. The phonon excitation spectrum is determined from the pole of this Green function which is given by the solution of the equation

$$\omega^2 - \omega_0^2 - 4\pi\omega_0\chi(q = 0, \omega) = 0. \tag{13}$$

The roots of this equation will show up peaks in the spectral density function. Replacing $\sum_k = \int d\varepsilon_k N(\varepsilon_k)$ and substituting the values of Γ functions from eqs (9a) to (9d) the final form of χ can be written as

$$\chi(q = 0, \omega) = (f_1^2(0)N(\varepsilon_F)/\pi) \int \left[[(\varepsilon_k - \tilde{E}'_0)^2 + 2\gamma_0(\varepsilon_k - \tilde{E}'_0)f_2(0)/f_1(0)\right.$$

$$\left. + (f_2^2(0)/f_1^2(0))\gamma_0^2] \frac{\tanh(y_2/2\theta) - \tanh(y_1/2\theta)}{(|D|(y_1 - y_2))} \right] d\varepsilon_k, \tag{14}$$

where

$$\left(\frac{1}{(e^{y_1/\theta} + 1)} - \frac{1}{(e^{y_2/\theta} + 1)} \right) = (1/2)\big(\tanh(y_2/2\theta) - \tanh(y_1/2\theta)\big). \tag{15}$$

It is evident from eq. (14) that $\chi(q = 0, \omega)$ is zero for $T = 0$. This is because, in this limit both $\tanh(y_1/2\theta)$ and $\tanh(y_2/2\theta)$ become equal to 1. However, on performing a low temperature expansion, eq. (15) can be expressed as

$$\tanh(y_2/2\theta) - \tanh(y_1/2\theta) = -4e^{-(\varepsilon_k + \bar{E}'_0)/2kT}$$

$$\times \sinh\sqrt{((\varepsilon_k - \tilde{E}'_0)^2 + 4\gamma_0^2)/2kT}. \tag{16}$$

The energy integration is taken over the conduction band width W from $-W/2$ to $W/2$, in the numerical claculation of the phonon energy. In the static limit ($c = \omega/\gamma_0 = 0$), the phonon self-energy can be expressed as

$$\chi(q=0,\omega) = -4(f_1^2(0)N(0)/\pi) \int (I_1 + I_2 + I_3) \left(\mathrm{e}^{-(\varepsilon_k + \bar{E}_0')/2kT} \right)$$

$$\times \left(\sinh \sqrt{((\varepsilon_k - \tilde{E}_0')^2 + 4\gamma_0^2/2kT} \right) \mathrm{d}\varepsilon_k, \tag{17}$$

where

$$I_1 + I_2 + I_3 = (\varepsilon_k - \tilde{E}_0')^2 + 2\gamma_0(\varepsilon_k - \tilde{E}_0')(f_2(0)/f_1(0))$$

$$+ (f_2^2(0)/f_1^2(0))\gamma_0^2/|D_1|(y_1 - y_2), \tag{18}$$

where $|D_1|$ is the value of $|D|$ in the static limit ($\omega = 0$) i.e.

$$|D_1| = (\varepsilon_k - E_0')(\varepsilon_k + E_0') - 4\gamma_0^2 - (U/2)\langle n_{k-\sigma}^f \rangle (\varepsilon_k - E_0'). \tag{19}$$

Moreover, the renormalized phonon frequenices in the static long wavelength limit can be expressed as

$$(\omega/\omega_0)^2 = 1 + (4\pi/\omega_0)\chi(q=0, \omega=0). \tag{20}$$

The phonon frequencies thus calculated will acquire a temperature dependence through eq. (17) besides depending on the various parameters such as the electron–phonon coupling constants and the intratomic Coulomb repulsion.

3. Results and discussions

It is well-known fact that the position of the f level relative to the Fermi level and the strength of hybridization plays an important role in HF systems. To understand in detail the behaviour of the elastic properties of these systems, we have investigated how the different parameters influence the phonon frequencies. The different dimensionless parameters that are involved in these calculations are the ratio of the two electron–phonon interaction strengths $r = f_2(0)/f_1(0)$; the dimensionless coupling constant $g = N(0)f_1^2(0)/\omega_0$, $N(0)$ being the density of states at the Fermi level. All the energies in the system are measured with respect to the strength of the hybridization (γ_0) which is the single dominant parameter, e.g., the position of the f-level is given by $d = E_0'/\gamma_0$ the onsite Coulomb repulsion $U' = U/\gamma_0$, the band width $W' = W/\gamma_0$ and the inverse of the temperature by $b' = \gamma_0/2KT$. Similarly the variable band energies are denoted by $y = \varepsilon_k/\gamma_0$ and the renormalized phonon frequency $\tilde{\omega} = \omega/\omega_0$ is measured with respect to the frequency (ω_0) of the bare phonon. Moreover, as per the finding of Razafimandiby *et al* [16], the value of r is always kept less than one so as to make $f_1(0)$ more stronger than $f_2(0)$). For simplicity, it is assumed that the f-level is half filled so that the average value of $\langle n_0^f \rangle$ is taken to be one. Since the Fermi level is set equal to zero ($E_f = 0$) the value of d is negative, if the f-level lies below the Fermi level, and is positive if it is above the Fermi level. The initial parameters were set by considering different physically allowed values of these at random for which uniform softening of phonon energy occurs.

The results of the numerical calculations are presented in four figures out of which figure 4 show the effect of Coulomb correlation parameter U on phonon frequency shifts.

In the first three figures the variation of phonon frequency with temperature is shown for different values of the parameters while the Coulomb correlation $U' = 0$. In the first three figures (figures 1–3), the variation of the reduced phonon frequency $\tilde{\omega}\,(= \omega/\omega_0)$ with inverse temperature b is plotted for different values of the parameters r, g and d while $U' = 0$. Figure 1 shows the variation of $\tilde{\omega}$ with temperature for different values of r keeping the other parameters g and d fixed at values 0.003 and -4.0 respectively. There is a reduction in phonon softening, when the values of r change from 0.1 to 0.001. Further it is observed that reduction of r beyond 0.005, hardly produces any noticeable effect on the phonon energy, e.g., for values of $r = 0.005$ and 0.001 the two curves coincide with only a slight change from that of the variation for $r = 0.01$. Figure 2 shows the variation of the reduced phonon frequency with temperature for different values of g (the effective coupling parameter) keeping r and d fixed at 0.01 and -4.0 respectively. From this figure, it is evident that, if g values are decreased from 0.05 to 0.0001, the softening of the phonon frequency also decreases. Small value of g means small value of $f_1^2(0)$ which implies that the coupling between phonons and hybridization of f and conduction electrons is weak and the strength is not sufficient to affect phonons. This is also in confirmation with the variation of r in figure 1. Figure 3 shows the variation with d (i.e. the position of f-level) while the other two parameters r and g are kept fixed at 0.01, 0.003 respectively. It is seen that, for negative values of d i.e. -6.0, -4.0 and -2.0, the phonon softening is rather
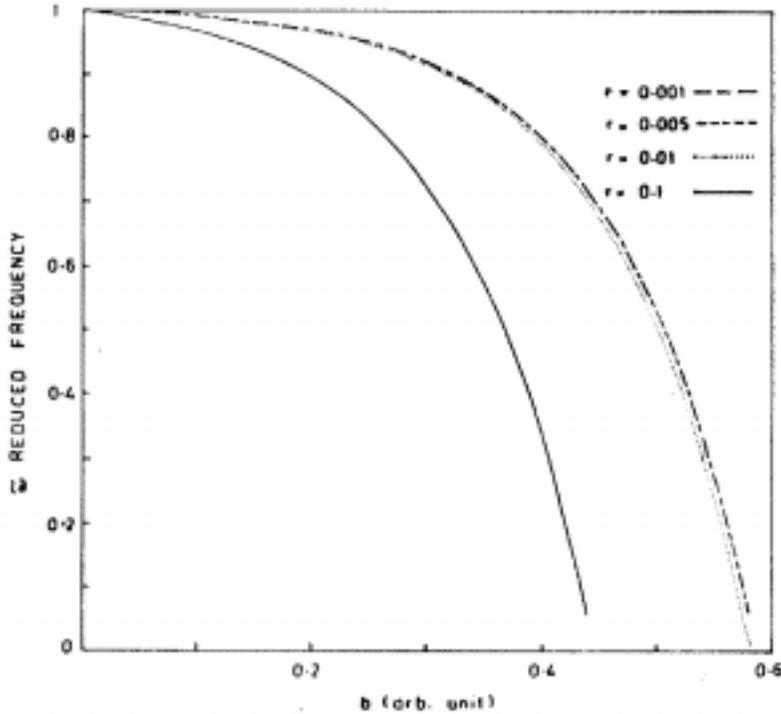

Figure 1. Plot of $\tilde{\omega}\,(= \omega/\omega_0)$ versus temperature b for $r = 0.001, 0.005, 0.01$ and 0.1 for fixed values of $g = 0.003$ and $d = -4$.
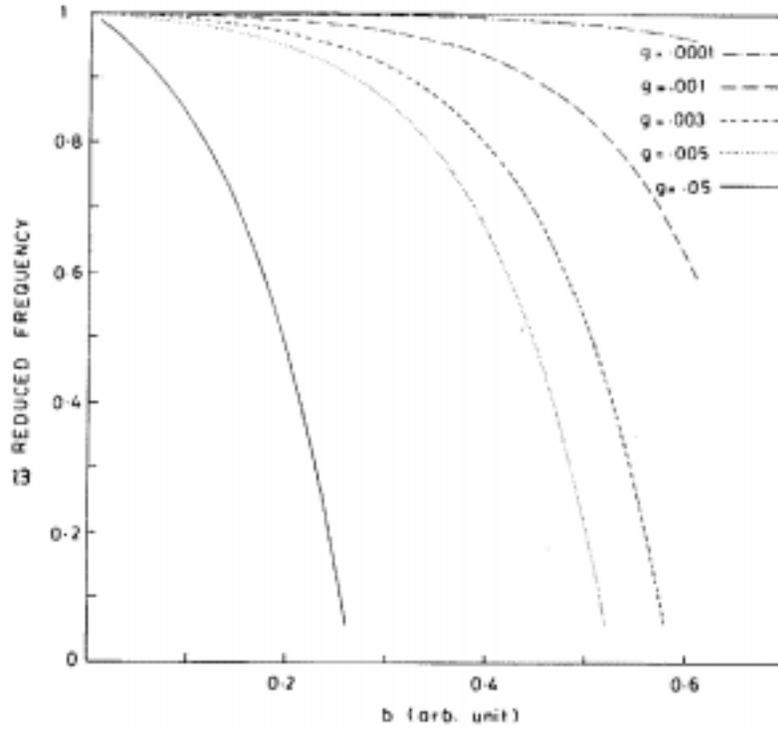
Figure 2. Plot of $\tilde{\omega}\, (= \omega/\omega_0)$ versus temperature b for $g = 0.0001$, 0.001, 0.003, 0.005 and 0.05 for fixed values of $r = 0.01$ and $d = -4$.

large. Moreover, as the f-level moves closer to conduction band i.e. going from -6 to -2 the degree of phonon softening decreases and covers a wider range of temperature. But for positive value of d the phonon softening is negligible. In all these plots increase of the parameter b implies the decrease in temperature as b is an inverse function of temperature.

In figure 4, we have shown the effect of correlation U on phonon softening. To visualize this effect, we have studied the variation of phonon energy with temperature considering the same initial values of the parameter r, g and d but for different values of U' including $U' = 0$. On comparison of the different plots, it is found that, as the value of U' increases the softening decreases. From the present theoretical analysis it is evident that the correlation shifts the position of the f-level. In the presence of correlation the bare f-level position E_0' becomes $\tilde{E}_0'\,(= E_0' + (U'/2)\langle n_{k,-\sigma}^f \rangle$ or $\tilde{d} = d + (U'/2)\langle n_{k,-\sigma}^f \rangle$. As the value of U increases from $U' = 0$ to some positive values, the value of the parameter d becomes less negative i.e. the f-level moves towards the Fermi level. From figure 3, it is evident that for the given values of r, g, d, the phonon softening decreases as the value of d is increased. Thus increase in correlation leads to hardening of phonons in agreement with the prediction of Min *et al* [14] and Hirsch [13].
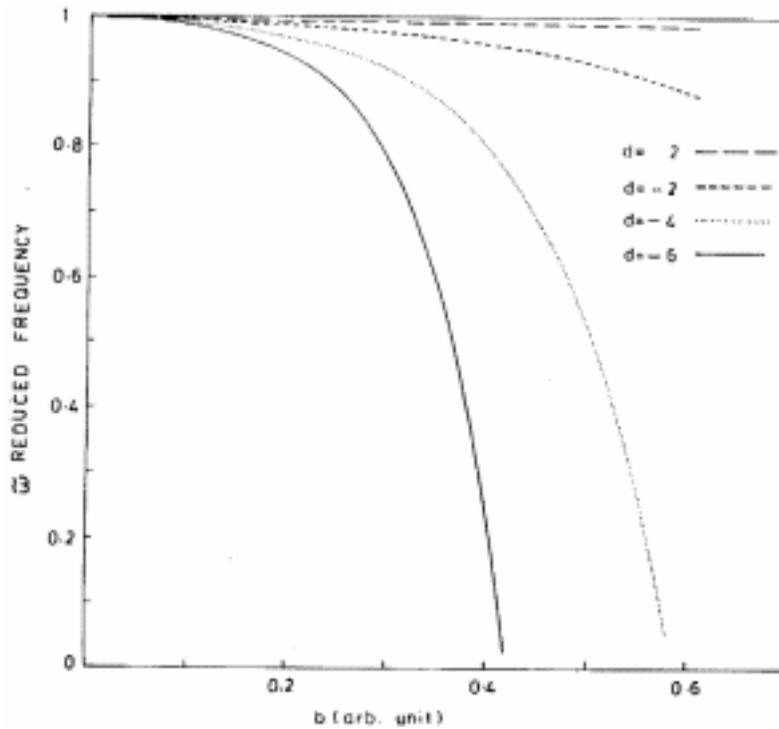
Figure 3. Plot of $\tilde{\omega}$ $(= \omega/\omega_0)$ versus temperature b for $d = -6$, -4, -2, and 2 for fixed values of $r = 0.01$ and $g = 0.003$.

4. Conclusion

In this section, we summarize the main results presented in this paper. An attempt has been made to explain the effect of the correlation on the electron–phonon interactions in the HF systems which indirectly influences the different physical properites exhibiting phonon anomalies. To understand this mechanism microscopically, phonons are assumed to interact with both the f-electrons as well as to the hybridization of conduction and f-electrons. Moreover, to study the different phonon anomalies exhbited by these systems, it is required to calculate the renormalized phonon frequencies. As a first step, we have calculated the phonon self-energy treating the Coulomb correlation between f-electrons with a mean field approximation. Zubarev [15] type Green functions were used to evaluate these quantities. The detail calculations of these quantities involve different electron response functions of f and conduction electron mixing as well as that of f-electrons and conduction electrons alone. The phonon response functions were evaluated exactly for all wave vectors (q) and temperatures (T). The Hubbard type of decoupling scheme [17] is carried out to reduce the higher order Green functions to the lower ones. To avoid complications, the numerical evaluations were made for $q = 0$ and in the limit of low temperatures. To achieve further simplification, the phonon self-energy is evaluated in the static limit keeping terms linear in U'. As per the procedure cited in the text, the initial fixed values of
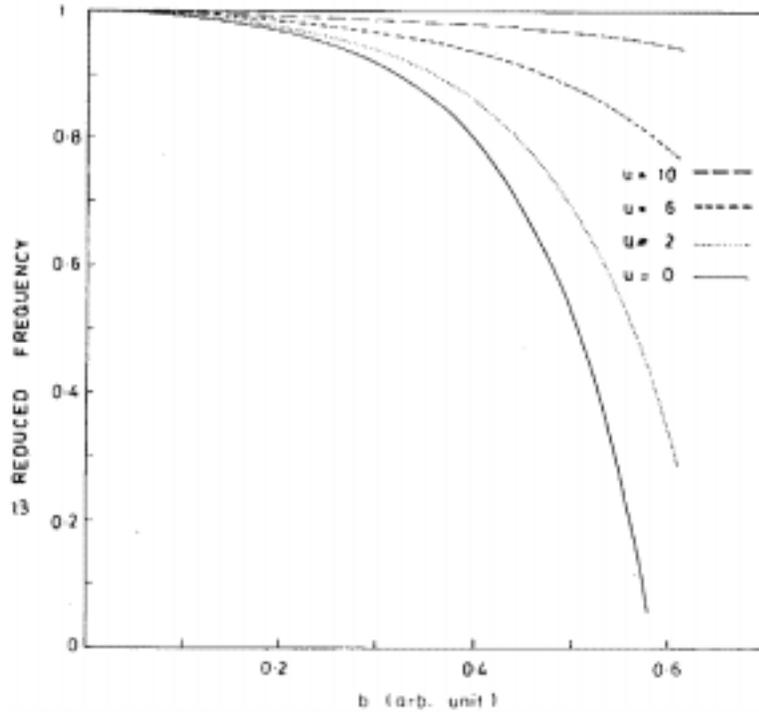
Figure 4. Plot of $\tilde{\omega}\,(= \omega/\omega_0)$ versus temperature b for $U' = 0, 2, 6$ and 10 for fixed values of $r = 0.01$, $g = 0.003$ and $d = -4$.

the important parameters r, g and d were found to be 0.01, 0.003 and -4.0 respectively. Considering these values along with different values of the correlation parameter U' we have shown that the influence of correlation is to shift the f-level closer to the Fermi level, thereby reducing the phonon softening.

The calculations presented in this paper can be improved by accounting for the effect of both finite q and T and considering these calculations in the dynamic limit. The calculation for finite q and T for $U' = 0$ is already reported [18]. However, the same calculation for finite U' is in progress and will be reported in future publications.

Acknowledgement

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