

Theoretical study of the Role of Strength of Hybridization on Specific heat of F-electron Systems

A K. SHDANGI¹ and G C ROUT^{2*}

¹ Ramajee Mahavidyalaya, Bhismagiri-761055, Odisha, India.

² Condensed Matter Physics Group, Physics Enclave, Plot No.- 664/4825, Lane 4A, Shree Vihar, CS. Pur, Bhubaneswar- 751031, Odisha, India

*Corresponding author, E-mail: gcr@iopb.res.in , asitkumar91519@gmail.com, E-mail¹: asitshad@iopb.res.in

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Abstract. f-electron systems are the compounds based on rare earth and actinide elements having partly filled f-electron orbitals, which below a characteristic low temperature, display very high specific heat coefficient with high effective electronic mass. In the present communication, we report a theoretical study of the role of the strength of hybridization between conduction and f-electrons on specific heat in these systems. We consider periodic Anderson model with Coulomb interaction within mean-field approximation and employ Zubarev's Green's function technique to calculate conduction electron as well as f-electron Green's function. The entropy, electronic specific heat and electronic specific heat coefficient are calculated from the electron free-energy of the f-electron system and are computed numerically.

Keywords. f-electron systems, Entropy and specific heat

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1. Introduction

Compounds containing rare earth and actinide elements having partly filled f-orbitals (f-electron systems), below a characteristic low temperature (T^*) exhibit many anomalous physical properties with high values of Sommerfeld coefficient (γ) and large electronic mass. For example, some Cerium based compounds such as $CeAl_3$ (1620 mJ/molK^2 at $T^*= 0.2\text{K}$), $CeCu_6$ ($\gamma = 1450 \text{ mJ/molK}^2$, $T^*= 4\text{K}$), $CeCu_2Si_2$ ($\gamma = 1100 \text{ mJ/molK}^2$, $T^*= 10\text{K}$), $CeRu_2Si_2$ ($\gamma = 385 \text{ mJ/molK}^2$, $T^*= 15\text{K}$) [1]. $CePt_2Sn_2$ ($\gamma = 350 \text{ mJ/molK}^2$, $T^*= 0.88\text{K}$) [2], $CePt_2Si_2$ ($\gamma = 170 \text{ mJ/molK}^2$, $T^*= 1.25\text{K}$) [3]

and $CePb_3$ ($\gamma > 350 \text{ mJ/molK}^2$, $T^* = 1.1\text{K}$) [4] are having high Sommerfeld coefficient and low fluctuation temperature. The actinide based compounds having low fluctuation temperature and high specific heat coefficient are UCd_{11} ($\gamma = \frac{\text{mJ}}{\text{molK}^2}$, $T^* = 5\text{K}$), $NpBe_{13}$ ($\gamma = 900 \text{ mJ/molK}^2$, $T^* = 3.4\text{K}$) [5]. UPt_3 ($\gamma = 450 \text{ mJ/molK}^2$ at 5.5K) [6] and URu_2Si_2 ($\gamma = 180 \text{ mJ/molK}^2$ at 17.5K) [7] both exhibit intermediate fluctuation temperature T^* i.e. 20K and 70K with intermediate specific heat coefficients 400 and 65 mJ/molK^2 respectively. Compound with high fluctuation temperatures are $T^* = 260\text{K}$ for $CeSn_3$ [8] and $T^* = 340\text{K}$ for $CeBe_{13}$ [9] and U_2R_2Sn [10, 11] are in mixed valence state. The temperature dependence of $\gamma(T)$ of f-electron systems like $CeAl_3$, $CeCu_6$ etc. increases with increase of temperature in the low temperature Fermi liquid or coherence regime, reaching a maximum (showing a peak) and then decreases exponentially with the increase of temperature [12, 13]. The large electronic mass and hence the high value of Sommerfeld specific heat coefficient is usually believed to be the result of the strong correlation between the localized f moments and the conduction electrons. Earlier, Rout and co-workers have studied ultrasonic attenuation, velocity of sound and the interplay of Kondo effect and f-electron correlation in heavy fermion systems and specific heat anomaly in heavy fermion superconductor $NdCuO_4$ [14-21].

In the present study, we have considered periodic Anderson model with Coulomb interaction within mean-field approximation. We have calculated conduction electron as well as f-electron Green's function by Zubarev's Green's function technique. The entropy, electronic specific heat and electronic specific heat coefficient are calculated from the electron free-energy and computed numerically.

2. Formalism

The f-electron system is described by the periodic Anderson model by the Hamiltonian which is written as

$$H_0 = \sum_{k,\sigma} (\varepsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k,\sigma} \varepsilon_f f_{k\sigma}^\dagger f_{k\sigma} + V \sum_{k,\sigma} (f_{k,\sigma}^\dagger c_{k,\sigma} + c_{k,\sigma}^\dagger f_{k,\sigma}) + U \sum_{k,\sigma} n_{i\sigma}^f n_{i-\sigma}^f \quad (1)$$

Here $c_{k\sigma}^\dagger$ ($c_{k\sigma}$) is the creation(annihilation) operator of conduction electrons and $f_{k\sigma}^\dagger$ ($f_{k\sigma}$) is the creation(annihilation) operator of f-electrons. ε_k and ε_f are conduction and f-electron energies. μ is the chemical potential. The third term describes the hybridization between conduction and f-electrons with V as

strength of hybridization. The fourth term describes Coulomb interaction between f- electrons

Using mean-field approximation, the Hamiltonian is rewritten as

$$H_0 = \sum_{k,\sigma} (\varepsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k,\sigma} E_0 f_{k\sigma}^\dagger f_{k\sigma} + V \sum_{k,\sigma} (f_{k,\sigma}^\dagger c_{k,\sigma} + c_{k,\sigma}^\dagger f_{k,\sigma}) \quad (2)$$

The Green's functions

$A_1(k, \omega) = \langle\langle c_{k\sigma}; c_{k\sigma}^\dagger \rangle\rangle_\omega$, $A_2(k, \omega) = \langle\langle f_{k\sigma}; c_{k\sigma}^\dagger \rangle\rangle_\omega$, and $F_1(k, \omega) = \langle\langle f_{k\sigma}; f_{k\sigma}^\dagger \rangle\rangle_\omega$ are calculated by equation of motion technique of Zubarev and are expressed as

$$A_1(k, \omega) = (\omega - E_0) / 2\pi |D(\omega)|, \quad A_2(k, \omega) = V/2\pi |D(\omega)| \text{ and} \\ F_1(k, \omega) = (\omega - \varepsilon_k) / 2\pi |D(\omega)| \text{ where, } |D(\omega)| = [\omega^2 - \omega(\varepsilon_k + E_0)^2 + \varepsilon_k E_0 - 4V^2].$$

Equating $|D(\omega)| = 0$, the quasi particle energies are found to be

$$\omega_1, \omega_2 = \frac{(\varepsilon_k + E_0) \pm \sqrt{(\varepsilon_k - E_0)^2 + 4V^2}}{2} \quad (3)$$

The f-electron occupation number determined self-consistently is

$$n^f = N_f(0) \int d\varepsilon_k \frac{1}{(\omega_1 - \omega_2)} [(\omega_1 - \varepsilon_k) f(\beta\omega_1) - (\omega_2 - \varepsilon_k) f(\beta\omega_2)] \quad (4)$$

3. Calculation of Entropy and Specific heat

The electronic free energy of the system is given by

$$F = -k_B T \sum_{k,\sigma} \ln[1 + \exp(-\beta\omega_\alpha)] \text{ where } \alpha = 1, 2, \quad (5)$$

where k_B is the Boltzmann constant.

The entropy of the system is defined as

$$S = -\left(\frac{\partial F}{\partial T}\right)_{V,\mu} \quad (6)$$

The specific heat is computed using the formula

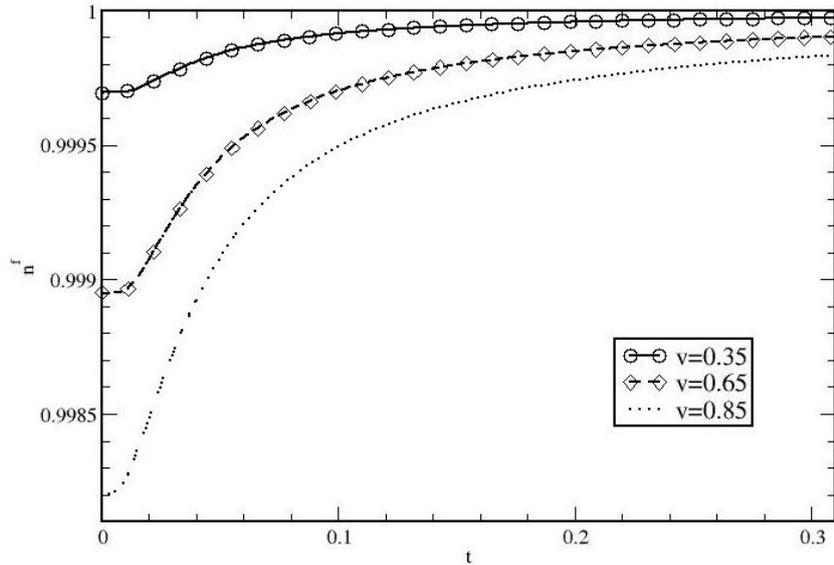
$$\frac{C_V}{k_B} = -T \left(\frac{\partial S}{\partial(k_B T)}\right)_{V,\mu} \quad (7)$$

The dimensionless parameters are the position of the bare f-level, $= \frac{\varepsilon_f}{\omega_D}$, the Coulomb energy, $u = \frac{U}{\omega_D}$, the strength of hybridization, $v = \frac{V}{\omega_D}$, chemical potential, $u_m = \frac{\mu}{\omega_D}$, the reduced temperature, $t = \frac{k_B T}{\omega_D}$, the entropy $s = \frac{S}{\omega_D}$, and $c_v = \frac{C_V}{\omega_D}$.

4. Results and Discussion

The f-electron occupancy i.e. n^f is calculated from the mean-field electron Hamiltonian given in equation (2) and is solved numerically and self-consistently for different values of position of f-electron(d) and Coulomb energy(u). The plots of f-electron occupancy (n^f) vs. temperature (t) are shown in Fig. 1.

Figure 1. Plot of f-electron occupancy (n^f) vs. temperature (t) for different values of



strength of hybridization $v = 0.35, 0.65, 0.85$ with fixed values of f-level energy $d = -0.94$ and Coulomb correlation energy $u = 1.0$.

The f-electron occupancy increases slowly and attains its integer value for lower strength of hybridization ($v = 0.35$), indicating the heavy fermion behavior of the system. The strength of hybridization between localized f electrons and conduction electrons becomes weak in the heavy fermion state as the position of f-level is away from the Fermi level. The system exhibits mixed valence behavior for stronger hybridization i.e. for $v = 0.65$ and $v = 0.85$ as the position of f-level becomes closer to the Fermi level and n^f moves away from its integer value and f-electron occupancy becomes less than 1.

The effect of hybridization strength on temperature dependent entropy is shown in Fig.2. The entropy of the system reduces in low temperature region, below the characteristic temperature, due to hybridization of f-electrons with conduction electrons. With the increase of hybridization strength from $v = 0.35$

to $\nu = 0.85$, the entropy of the system enhances sharply indicating that the system moves from heavy fermion state to mixed valence state

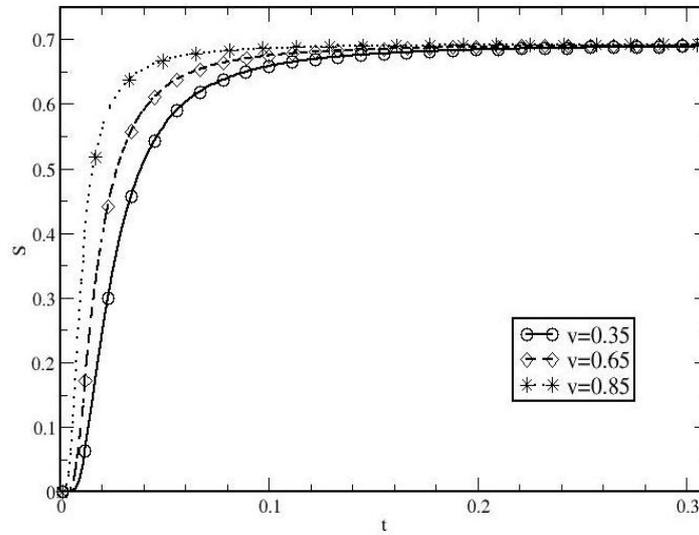


Figure 2. Plot of entropy(S) vs. temperature (t) for different values of strength of hybridization $\nu = 0.35, 0.65, 0.85$ with fixed values of f-level energy $d = -0.94$ and Coulomb correlation energy $u = 1.0$

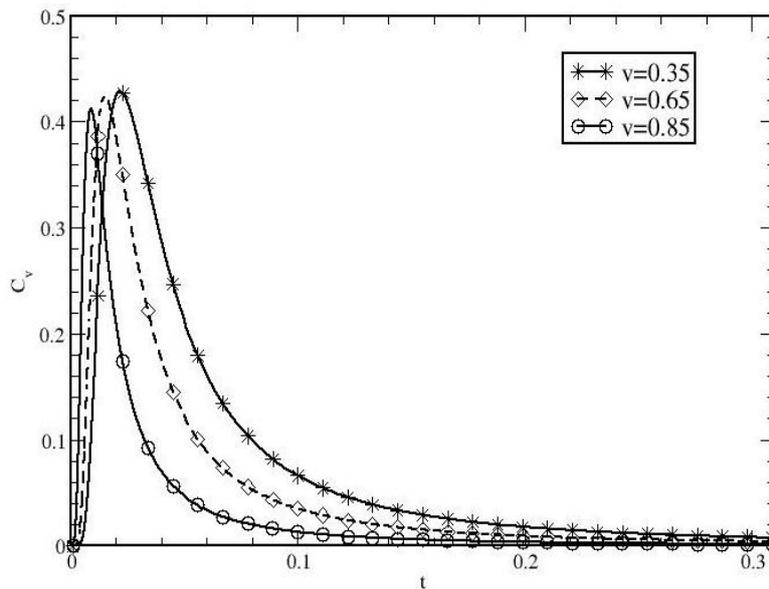


Figure 3. Plot of specific heat vs. temperature (t) for different values of strength of hybridization $\nu = 0.35, 0.65, 0.85$ with fixed values of f-level energy $d = -0.94$ and Coulomb correlation energy $u = 1.0$.

The temperature dependent electron specific heat is shown Fig.3. At low temperature, the specific heat rises linearly to its peak value at the characteristic temperature $t^* = 0.025$ showing the heavy fermion character and then it decreases exponentially with increase of temperature. As the strength of hybridization increases, the peak position in C_v shifts to lower temperature with slight decrease in peak height. Hence, increase of strength of hybridization suppresses the heavy fermion behavior of the system.

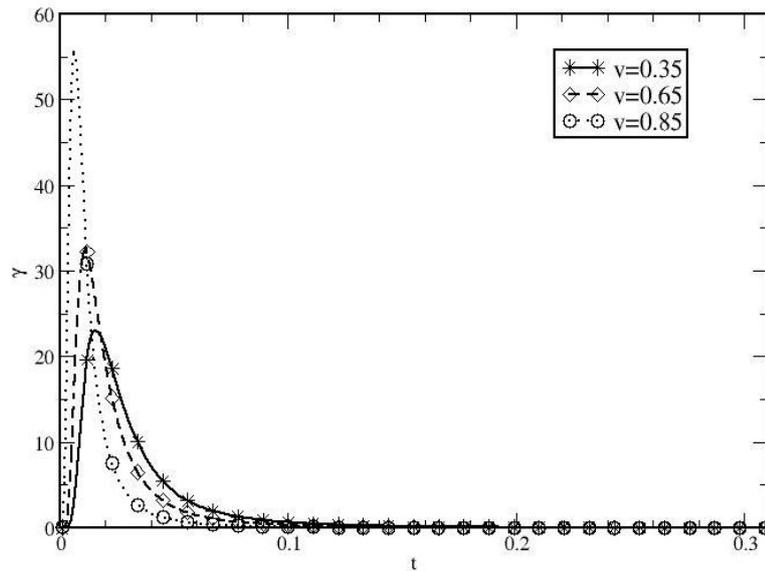


Figure 4. Plot of specific heat coefficient (γ) vs. temperature (t) for different values of strength of hybridization $v = 0.35, 0.65, 0.85$ with fixed values of f -level energy $d = -0.94$ and Coulomb correlation energy $u = 1.0$.

The plot of temperature dependent linear specific heat coefficient γ for different values of strength of hybridization $v = 0.35, 0.65$ and 0.85 is shown in fig.4. For lower $v = 0.35$, γ increases with temperature, attains peak and then falls with increase of temperature. For further increase of strength of hybridization, the peak in specific heat coefficient γ becomes sharper and shifts to lower characteristic temperature along with enhancement of peak heights. It is observed that, the high value of γ is associated with lower characteristic temperature and the vice versa [1-8]. The electrons of $5f$ orbitals of actinides (like U and Np) are less localized than the $4f$ electrons of rare earth (like Ce, Yb) systems and hybridize strongly with conduction electrons. The systems with

actinide elements show very high specific heat coefficient at low characteristic temperature.

There is often an intermediate state between localized and delocalized 5f electrons in many actinide compounds. Thus it is concluded that the rare earth systems will have higher linear specific heat coefficient as compared to that of Uranium based systems.

4. Conclusion

We have considered the well-known periodic Anderson model taking Coulomb interaction within Hartree-Fock approximation. We have calculated the f-electron occupation number, entropy, specific heat as well as specific heat coefficient for f-electron systems. For different strength of hybridization, the entropy of the system reduces and specific heat and Sommerfeld coefficient attains peak at the different low characteristic temperature as observed in different rare earth and actinide based compounds. Hence, the strength of hybridization between conduction and f-electrons is found playing a crucial role behind the low temperature anomalies in specific heat of f-electron systems.

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