

A study on local moments and charge fluctuation in mixed-valence systems

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Abstract : An attempt is made to study local moments and charge fluctuation in mixed-valence systems. Model Hamiltonian approach has been accepted. Role of f - f Coulomb interaction and electron-phonon (EP) interaction is investigated. A comparison is also made with few existing results.

Keywords : Mixed-valence systems, local moments and charge fluctuation, Coulomb interaction, electron-phonon interaction

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1. Introduction

Plenty of studies on mixed-valence (MV) phenomena exhibited by rare-earth intermetallics and compounds have been done using various model Hamiltonians [1-3]. Recently, these model Hamiltonians have been successfully used to explain the characteristic features of local moments and charge fluctuations which are observed in the majority of rare-earth materials [4-6].

Blankenbecler *et al* [7] considered a one dimensional, symmetric, periodic Anderson model and studied the ground state properties using stochastic Monte-Carlo techniques. They have shown that the ground state exhibits short-range magnetic correlations and that the local f -electron spin moments are compensated by correlations with other f -electrons as well as band electrons leading to a nonmagnetic ground state. NIE and WEI [8] used the local approach to study the correlation effects of the periodic Anderson lattice. Their results showed that the Coulomb interaction U leads to a formation of local moments and a reduction of charge fluctuation. It is in agreement with Yamada and Yosida [9] who investigated the ground-state properties of the periodic Anderson Hamiltonian using higher-order perturbation theory.

In the present work, the possible nature of local moments and charge fluctuation in MV systems has been investigated. We have considered a two-site model Hamiltonian [10], which incorporates both the Falicov-Kimball term and the electron-phonon (EP) interaction term. A suitable representative state is chosen [10] for which the exact calculation is

admissible. Effect of different interactions on local moments and charge fluctuation have been studied.

2. Formulation

We have considered the following model Hamiltonian :

$$H = H_0 + H_v \quad (1)$$

$$\begin{aligned} \text{where } H_0 = & \sum_{i,\sigma} (E_0 d_{i\sigma}^\dagger d_{i\sigma} + E f_{i\sigma}^\dagger f_{i\sigma}) + G \sum_{i,\sigma,\sigma'} d_{i\sigma}^\dagger d_{i\sigma} f_{i\sigma'}^\dagger f_{i\sigma'} \\ & + U_{ff} \sum_i f_{i\uparrow}^\dagger f_{i\uparrow} f_{i\downarrow}^\dagger f_{i\downarrow} + W \sum_{\substack{i,j,\sigma \\ i \neq j}} d_{i\sigma}^\dagger d_{j\sigma} \end{aligned} \quad (2)$$

$$\text{and, } H_v = V \sum_{\substack{i,j,\sigma \\ i \neq j}} (f_{i\sigma}^\dagger d_{j\sigma} + d_{j\sigma}^\dagger f_{i\sigma}) - g \sum_{\substack{i,j,\sigma,\sigma' \\ i \neq j}} n_{i\sigma}^d (f_{i\sigma}^\dagger d_{j\sigma} + d_{j\sigma}^\dagger f_{i\sigma}) \quad (3)$$

E_0, E are the d -level and f -level energy, respectively, G is the strength of the f - d Coulomb interaction, U_{ff} the Coulomb repulsion between the f -electrons of opposite spins on the same site, W the strength of the hopping interaction between d -electrons at different sites. V the f - d hybridisation parameter, and g is the electron-phonon interaction parameter. We have restricted ourselves to the case $\langle n_{i\uparrow}^f \rangle + \langle n_{i\downarrow}^f \rangle = 1$. Also, throughout our calculation $i, j = 1, 2$ and $\sigma, \sigma' = \uparrow, \downarrow$.

The representative two-site spin state is taken in the form

$$| n_{1\uparrow}^f n_{1\downarrow}^d n_{2\uparrow}^d n_{2\downarrow}^f n_{2\uparrow}^f n_{2\downarrow}^d n_{2\uparrow}^d n_{2\downarrow}^f \rangle \quad (4)$$

and the eigenvectors of H are represented as a linear combinations of vectors (4).

In such a way we have solved the eigen equation

$$H | E_\alpha \rangle = E_\alpha | E_\alpha \rangle, \quad \alpha = 1, \dots, 28. \quad (5)$$

The ground state $|\psi\rangle$ is constructed for which E_α has the lowest value.

The local moments S_m and the charge fluctuation Q_f are calculated using the following formulas,

$$\begin{aligned} S_m &= 3/4 (\langle n_i^f \rangle - 2 \langle n_{i\uparrow}^f n_{i\downarrow}^f \rangle), \\ Q_f &= \langle n_i^f \rangle (\langle n_i^f \rangle - 1) + 2 \langle n_{i\uparrow}^f n_{i\downarrow}^f \rangle. \end{aligned}$$

3. Results and discussions

Figure 1 shows the variation of the local moments S_m with U_{ff} for different hybridisation strengths. The local moment is increased with increasing U_{ff} for fixed V . The Coulomb interaction U_{ff} reduces the probability of finding two f -electrons with opposite spins at the same site, and leads to a formation of local moments on the f -orbitals. Since we are considering the system in the region of phase transition, the hybridisation V has two

opposing effects on f -electron occupation number in these two phases (insulating and metallic). As the value of U_{ff} increases, the system gradually enters into the metallic phase. $\langle n_i^f \rangle$ as well as $\langle n_{i\uparrow}^f n_{i\downarrow}^f \rangle$ decreases with V in the insulating phase and increases with V in the metallic phase. Finally, we obtain the characteristics of S_m as shown in the figure.

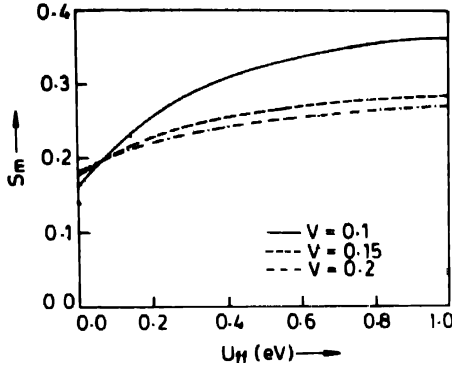


Figure 1. S_m as a function of U_{ff} for $T = 0$ K and for various values of V (in eV). $E_0 = 0$ eV, $E = -0.1$ eV, $G = 0.5$ eV, $W = 0.06$ eV, $g = 0$ eV

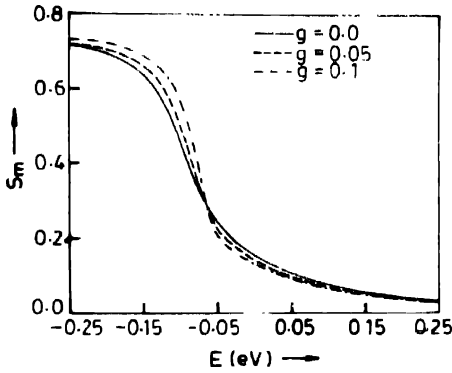


Figure 2. S_m as a function of E for $T = 0$ K and for various values of g (in eV). $E_0 = 0$ eV, $G = 0.25$ eV, $W = 0.06$ eV, $U_{ff} = 1.0$ eV, $V = 0.1$ eV.

S_m varies with E in the same way as $\langle n_i^f \rangle$ as is apparent in Figure 2, where local moment has been plotted against E for different EP interaction. As the value of E increases, $\langle n_i^f \rangle$ decreases which reduces the formation of local moments on the f -orbitals. The fluctuation of $\langle n_{i\uparrow}^f n_{i\downarrow}^f \rangle$ with E , and EP interaction is very small compared to $\langle n_i^f \rangle$ and so in the present case, $\langle n_i^f \rangle$ determines S_m . In the insulating phase, EP interaction increases S_m following $\langle n_i^f \rangle$. With the increase of E phase transition occurs and S_m decreases with g .

In Figure 3, we have plotted the charge fluctuation Q_f vs U_{ff} for different hybridisation strengths. The result shows that the charge fluctuation Q_f is decreased with increasing U_{ff} for fixed V . As the value of U_{ff} increases, $\langle n_i^f \rangle$ as well as $\langle n_{i\uparrow}^f n_{i\downarrow}^f \rangle$

decreases. These make Q_f decrease with U_{ff} . The result is in agreement with local approach method in periodic Anderson lattice [8]. Here also, in the insulating phase, Q_f decreases with V and in the metallic phase, Q_f increases with V .

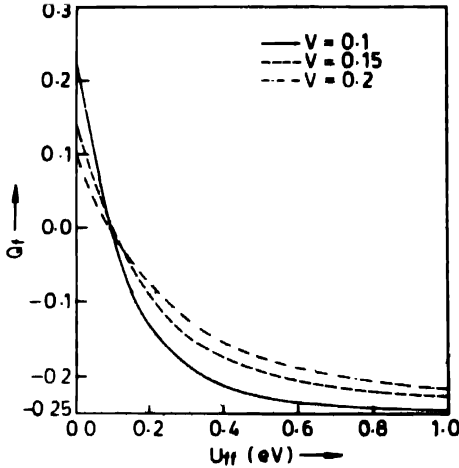


Figure 3. Q_f as a function of U_{ff} for $T = 0$ K and for various values of V (in eV). $E_0 = 0$ eV, $E = -0.1$ eV, $G = 0.5$ eV, $W = 0.06$ eV, $g = 0$ eV.

Curves of the charge fluctuation against E are presented in Figure 4 from which we find that with the increase of E , Q_f decreases from a value close to zero and after a certain E , it increases towards the value zero. Here, as charge fluctuation is primarily determined by $\langle n_i^f \rangle$, so, in the insulating or metallic phase the charge fluctuation should approach zero. For large negative E , EP interaction keeps the value of $\langle n_i^f \rangle$ close to unity, and after the transition keeps a value of $\langle n_i^f \rangle$ close to zero. In both the cases Q_f should be closer to zero than that for smaller EP interaction as is apparent in Figure 4.

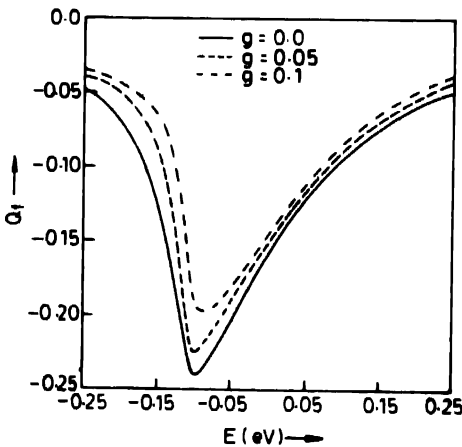


Figure 4. Q_f as a function of E for $T = 0$ K and for various values of g (in eV). $E_0 = 0$ eV, $G = 0.25$ eV, $W = 0.06$ eV, $U_{ff} = 1.0$ eV, $V = 0.1$ eV.

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