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# QUASI PARTICLE ENERGY OF 4f-STATES IN THE RAMIREZ-FALICOV-KIMBALL (RFK) MODEL: MEMORY FUNCTION FORMALISM

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**Abstract.** - A new formalism is developed, based on the memory function approach, to treat many particle systems. The formalism is applied to the Ramirez-Falicov-Kimball (RFK) Hamiltonian, suitable to describe photoemission spectra in many light rare earth intermetallics. We obtain a quasi particle 4f-energy in the weak correlation regime and we discuss the bimodal structure of the f-f propagator in this regime comparing with the Hubbard-type structure in the strong correlation regime.

It is well known that many experiments concerning the photo-emission of 4f-electrons in light rare-earth elements, e.g., Ce, show a double peak structure: one localized at the Fermi level and another approximately 2.5 eV below it.

Parks *et al.* [1] and Wieliczka *et al.* [2] have shown that this bimodal structure of the 4f-spectra occurs in many other metallic systems containing light rare earths such as Pr and Nd.

Many works [3, 4, 5] have been proposed in order to explain the 4f-double structure, based, for example, on the rare earth magnetic properties [3] or on screening effects [4, 5]. Nunez-Regueiro and Avignon [6] have calculated the 4f-spectral density, based on the Falicov-Kimball model, adopting Hubbard's "resonance broadening approximation" . This strong correlation regime approximation, yields one or two peaks depending on the ratio between the Coulomb correlation $U$ between the f-localized states and the d-itinerant states and the d-bandwidth $\Delta$. Moreover, f-d hybridization plays no significant role in the broadening of the two peaks.

In this work, adopting the Ramirez-Falicov-Kimball (RFK) Hamiltonian, we calculate the f-f Green's function in the weak correlation regime, i.e., $U/W < 1$. We develop here a Memory Function matrix formalism, which enables us to describe the weak correlation regime beyond the usual Hartree-Fock approximation.

For the sake of simplicity, we discuss here only the RFK Hamiltonian in the one-impurity case:

$$\begin{aligned} H = & \sum_{\sigma} \varepsilon_0 \, f_{0\sigma}^{+} \, f_{0\sigma} + \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} \, d_{\mathbf{k}\sigma}^{+} \, d_{\mathbf{k}\sigma} + \\ & + \sum_{\mathbf{k}\sigma} V \left( F_{0\sigma}^{+} \, d_{\mathbf{k}\sigma} + d_{\mathbf{k}\sigma}^{+} \, f_{0\sigma} \right) + \sum_{\sigma\sigma'} U n_{0\sigma}^{\mathrm{d}} \, n_{0\sigma'}^{\mathrm{f}} \; ; \\ & n_{0\sigma}^{\alpha} = \alpha_{0\sigma}^{+} \, \alpha_{0\sigma'} \; ; \\ & \sum_{\sigma} n_{0\sigma}^{\alpha} = n_0^{\alpha} \; , \quad (\alpha = f \text{ or d}). \end{aligned} \tag{1}$$

The local f-f Green function is given by

$$G_{00\sigma}^{\mathrm{ff}}(t) = i\theta(t) \left\langle \left[ f_{0\sigma}, \, f_{0\sigma}^{+}(t) \right]_{+} \right\rangle . \tag{2}$$

Now we introduce the self-consistent many body theory developed by Fedro and Wilson [7], Kishore [8] and Chao *et al.* [9]. Let us consider two sets of Heisenberg fermion operators $A_\alpha$ and $B_\beta$ forming a complate space:

$$\begin{aligned} \{A_\alpha\} &= \left\{ f_{0\sigma}, \, d_{\mathbf{k}\sigma} \right\} \\ \{B_\beta\} &= \left\{ f_{0\sigma}^{+}, \, d_{\mathbf{k}\sigma}^{+} \right\} \end{aligned} \tag{3}$$

and a projection operator $P$ defined as

$$P\Psi = \sum_j P_j \, \Psi = \sum_j B_j \frac{\left\langle [A_j, \, \Psi]_+ \right\rangle}{\left\langle [A_j, \, B_j]_+ \right\rangle} . \tag{4}$$

Using the sets given by equation (3), we have:

$$P\Psi = f_{0\sigma}^{+} \left\langle |f_{0\sigma}, \, \Psi|_{+} \right\rangle + \sum_{\mathbf{k}} d_{\mathbf{k}\sigma}^{+} \left\langle \left[ d_{\mathbf{k}\sigma}, \, \Psi \right]_{+} \right\rangle . \tag{5}$$

An equation of motion for the matrix $\tilde{G}(w)$ :

$$G_{\alpha\beta}(t) = i\theta(t) \left\langle [A_\alpha, \, B_\beta(t)]_+ \right\rangle \tag{6}$$

can be worked out:

$$\tilde{G}(w) = \left[ x\tilde{I} - \tilde{\Omega} - \tilde{\gamma}(w) \right]^{-1} \tilde{\chi} \tag{7}$$

where

$$\Omega_{\alpha\beta} = \frac{\left\langle [A_\alpha \, LB_\beta]_+ \right\rangle}{\left\langle [A_\alpha, \, B_\alpha]_+ \right\rangle} \tag{8}$$

$$\chi_{\alpha\beta} = \left\langle [A_\alpha, \, B_\beta]_+ \right\rangle \delta_{\alpha\beta} \tag{9}$$

and

$$\gamma_{\alpha\beta}(w) = \left\langle \left[ A_\alpha, \, L \frac{1}{w - (1-P)L} (1-P) \, LB_\beta \right]_+ \right\rangle , \tag{10}$$

$L$ being the Liouvillean operators: $L\Psi \equiv [H, \, \Psi]$.

If we identify our first matrix element with the f-state, we have:

$$G_{00\sigma}^{\mathrm{ff}}(w) = \left[ w\tilde{I} - \tilde{\Omega} - \tilde{\gamma}(w) \right]_{11}^{-1} \chi_{11} . \tag{11}$$

Equation (11) can be solved in several levels of approximations for the matrix $\tilde{\gamma}(w)$. In the lowest level of approximation we use the linearized f-d Coulomb term in the Hamiltonian. Then we find: $\tilde{\gamma}(w) = 0$. The f-f propagator becomes:

$$G_{00\sigma}^{\mathrm{ff}}(w) = \frac{1}{w - \varepsilon_0^{\mathrm{f}} - U\left\langle n_0^{\mathrm{d}} \right\rangle - V^2 \, F(w)} \tag{12}$$

where

$$F(w) = \sum_{\mathbf{k}} \frac{1}{w - \varepsilon_{\mathbf{k}} - U\left\langle n_0^{\mathrm{f}} \right\rangle} . \tag{13}$$

and we recover the Hartree-Fock approximation.

In the next step, we use a recursion formula for the self-energy $\gamma(w)$ [9, 10].

The hierarchy of the Green's function is truncated by approximating conveniently the self-energy $\gamma^{\mathrm{ff}}(n+1 : w)$. Thus, in the first order approximation, we linearize the Hamiltonian for $\gamma^{\mathrm{ff}}(2 : w)$, which will give us again $\gamma^{\mathrm{ff}}(2 : w) = 0$. Then we obtain from the recursion formula:

$$\gamma^{\mathrm{ff}}(1 : w) = \frac{U^2 \langle n_0^{\mathrm{d}} \rangle \left(1 - \langle n_0^{\mathrm{d}} \rangle\right)}{w + \varepsilon_0 + U \langle n_0^{\mathrm{d}} \rangle} . \tag{14}$$

$G_{00\sigma}^{\mathrm{ff}}$ exhibits a bimodal structure in the weak correlation regime. This bimodal structure however is quite different from the Hubbard-type two-peak structure [6] which is peculiar to a strong correlation regime. The two resonant f-energies are:

$$E_{\pm} = \frac{V^2 F(w)}{2} \pm \frac{1}{2} \sqrt{\left[2\varepsilon_0 + 2U \langle n_0^{\mathrm{d}} \rangle + V^2 F(w)\right]^2 + 4U^2 \langle n_0^{\mathrm{d}} \rangle \left(1 - \langle n_0^{\mathrm{d}} \rangle\right)} . \tag{15}$$

The f-f propagator, exhibiting a $n$-modal structure is obtained by linearizing again the Coulomb interaction contribution for higher $\gamma^{\mathrm{ff}}(n+1 : w)$ terms in the recursion formula. As an illustration of this peculiar feature, we perform the calculation up to a higher level of approximation, truncating the expansion terms in $\gamma^{\mathrm{ff}}(3 : w)$, giving rise to terms in $U^3$. Then, we have:

$$\gamma^{\mathrm{ff}}(w) = \frac{\left(w \left\langle \left[f_{0\sigma}, L(1-P) L f_{0\sigma}^{+}\right]_{+} \right\rangle + \left\langle \left[f_{0\sigma}, L^2 (1-P) L f_{0\sigma}\right]_{+} \right\rangle\right)}{w^2 + w \left\langle [f_{0\sigma}, L f_{0\sigma}^{+}]_{+} \right\rangle + \left\langle [f_{o\sigma}, L^2 f_{0\sigma}^{+}]_{+} \right\rangle} \tag{16}$$

and after some algebra we obtain:

$$\gamma^{\mathrm{ff}}(w) = \frac{wU^2 \langle n_0^{\mathrm{d}} \rangle \left(1 - \langle n_0^{\mathrm{d}} \rangle\right) + U^2 \langle n_0^{\mathrm{d}} \rangle \left(1 - \langle n_0^{\mathrm{d}} \rangle\right) (2\varepsilon_0 + U) + V^2 U \left(\langle n_0^{\mathrm{f}} \rangle - \langle n_0^{\mathrm{d}} \rangle\right)}{w^2 + w \left(\varepsilon_0 + U \langle n_0^{\mathrm{d}} \rangle\right) + \left(\varepsilon_0^2 + 2\varepsilon_0 U \langle n_0^{\mathrm{d}} \rangle + U^2 \langle n_0^{\mathrm{d}} \rangle + V^2\right)} . \tag{17}$$

Introducing the above result in equation (11) the f-f Green functionwhich exhibits a tri-modal structure for the 4f-spectral density of states, associated to the higher order of the approximaton on the self-energy $\gamma^{\mathrm{ff}}(w)$.

If one goes further in our perturbative treatment one can obtain, in principle, a $n$-modal structure for the f-f propagator. However, for the physical situation which we are interested in, one needs only to go up to second order in $U$, where the main features of the 4f-states structures are already present (cf. Eq. (19)).

Finally, it should be mentioned, that this approach can also be applied in the case of strong correlation limit, i.e., $U / \Delta \gg 1$. In this case, the choice of the starting set of operators is a different one, namely:

$$\begin{aligned} \langle A_i^{+} \rangle &= \left\{ f_{0\sigma}\, n_0^{\mathrm{d}+},\ d_{\mathbf{k}\sigma} \right\} \\ \langle A_i^{-} \rangle &= \left\{ f_{0\sigma}\, n_0^{\mathrm{d}-},\ d_{\mathbf{k}\sigma} \right\} \\ \langle B_i \rangle &= \left\{ f_{o\sigma},\ d_{\mathbf{k}\sigma} \right\} \end{aligned} \tag{18}$$

where:

$$\begin{aligned} n_0^{\mathrm{d}+} &= n_0^{\mathrm{d}} , \\ n_0^{\mathrm{d}^-} &= 1 - n_0^{\mathrm{d}} . \end{aligned} \tag{19}$$

With this choice, the f-f propagator can be written as:

$${}^{\mathrm{ff}}_{00\sigma}(w) = G_{00\sigma}^{\mathrm{ff}+}(w) + G_{00\sigma}^{\mathrm{ff}-}(w) \tag{20}$$

where

$$G_{00\sigma}^{\mathrm{ff}\pm}(w) = i\theta(t) \left\langle \left[f_{0\sigma}\, n_0^{d\pm},\ f_{0\sigma}^{+}\right] \right\rangle . \tag{21}$$

In the lowest approximation and assuming $V = 0$ (i.e., a Falicov-Kimball model), one gets the usual Hubbard-type bimodal structure

$$G_{00\sigma}^{\mathrm{ff}}(w) = \frac{1 - \langle n_0^{\mathrm{d}} \rangle}{w - \varepsilon_0} + \frac{\langle n_0^{\mathrm{d}} \rangle}{w - \varepsilon_0 - U} , \tag{22}$$

which is completely different from the bimodal structure derived in this work, in the weak correlation regime.


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