



## Exact solution of the multicomponent Falicov–Kimball model in infinite dimensions

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### ABSTRACT

The exact solution for the thermodynamic and dynamic properties of the infinite-dimensional multicomponent Falicov–Kimball model for arbitrary concentrations of d and f electrons is presented. The emphasis is on a descriptive derivation of important physical quantities by the equation-of-motion technique. We provide a thorough discussion of the f-electron Green's function and of the susceptibility to spontaneous hybridization. The solutions are used to illustrate different physical systems ranging from the high-temperature phase of the YbInCu<sub>4</sub> family of materials to an examination of classical intermediate-valence systems that can develop a spontaneous hybridization at  $T = 0$ .

### §1. INTRODUCTION

The anomalous features observed in the YbInCu<sub>4</sub> family of intermetallic compounds (Sarrao *et al.* 1999) seem to be driven by the short-range Coulomb interaction between highly degenerate Yb f holes and the conduction states (Freericks and Zlatić 1998, Zlatić and Freericks 2001). The same interaction seems to be responsible for the optical anomalies in SmB<sub>6</sub> and related compounds (such as correlated ferroelectrics) (Guntherodt *et al.* 1982, Wachter and Travaglini 1985, Portengen *et al.* 1996a,b). We study the effect of this interaction using the multicomponent Falicov–Kimball (FK) (1969) model in infinite dimensions, where all the thermodynamic properties can be calculated exactly. The model consists of  $(2s + 1)$ -fold degenerate mobile d electrons and static  $(2S + 1)$ -fold degenerate f electrons, which interact via an on-site Coulomb interaction  $U$ . The model was originally introduced to describe metal–insulator transitions in materials that do not change the character of their electronic states but do change their thermodynamic occupations as functions of the external parameters. The exact results for the static and dynamic correlation functions of the spin- $\frac{1}{2}$  model explain the collapse of the low-temperature metallic phase of YbInCu<sub>4</sub>-like systems and account in a qualitative way for most of their features in the paramagnetic semiconducting high-temperature phase. The exact solution of

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the spinless model shows that the statistical fluctuations give rise to a logarithmic divergence (in  $T$ ) of the spontaneous hybridization correlation function at zero temperature, so that any amount of quantum mixing could lead to a phase transition at finite temperatures. This might be relevant for  $\text{SmB}_6$  and for correlated ferro-electrics (Portengen *et al.* 1996a,b).

In what follows, we describe the model, explain the method of calculating the Green's functions for d and f electrons and present results for some static and dynamic correlation functions of a spinless and spin-degenerate case. Detailed comparison with the experimental data will be given elsewhere.

## §2. FORMALISM FOR THE d-ELECTRON GREEN'S FUNCTION

The multicomponent FK model describes the dynamics of two types of electron: conduction electrons (created or destroyed at site  $i$  by  $d_{i\sigma}^\dagger$  or  $d_{i\sigma}$ ) and localized electrons (created or destroyed at site  $i$  by  $f_{i\eta}^\dagger$  or  $f_{i\eta}$ ). The  $(2s+1)$ -fold degenerate d states and the  $(2S+1)$ -fold degenerate f states are labelled by  $\sigma$  and  $\eta$  respectively. The multicomponent model is used to describe the electrons with spin and/or orbital degrees of freedom, and  $2s+1$  and  $2S+1$  can assume different values. The non-interacting conduction electrons can hop between nearest-neighbour sites on the  $D$ -dimensional lattice, with a hopping matrix  $-t_{ij} = -t^*/2D^{1/2}$ ; we choose a scaling of the hopping matrix that yields a non-trivial limit in infinite dimensions (Metzner and Vollhardt 1989). The f electrons have a site energy  $E_f$ , and a chemical potential  $\mu$  is employed to conserve the total number of electrons  $n_d + n_f = n_{\text{tot}}$ . The d- and f-number operators at each site are  $n_d = \sum_{\sigma} n_{d\sigma}$  and  $n_f = \sum_{\eta} n_{f\eta}$ . We assume an infinite Coulomb repulsion between f electrons with different labels  $\eta$  and restrict the f occupancy at a given site to  $n_f \leq 1$ , regardless of the degeneracy of the f state. The Coulomb interaction  $U$  between the d and f electrons that occupy the same lattice site is finite, and the FK Hamiltonian for the lattice is defined as (Brandt and Mielsch 1989, Freericks and Zlatić 1998, Zlatić and Freericks 2001)

$$H_{\text{FK}} = \sum_{ij,\sigma} (-t_{ij} - \mu\delta_{ij}) d_{i\sigma}^\dagger d_{j\sigma} + \sum_{i,\eta} (E_f - \mu) f_{i\eta}^\dagger f_{i\eta} + U \sum_{i,\sigma\eta} d_{i\sigma}^\dagger d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}. \quad (1)$$

The FK lattice model (1) can be solved in infinite dimensions using the methods of Brandt and Mielsch (1989). We consider two kinds of lattice: firstly, the hypercubic lattice with a Gaussian non-interacting density of states,  $\rho(\epsilon) = \exp(-\epsilon^2/t^{*2})/\pi t^*$ ; and, secondly, the infinite-coordination Bethe lattice with a semicircular non-interacting density of states,  $\rho(\epsilon) = (4t^{*2} - \epsilon^2)^{1/2}/(2\pi t^{*2})$ ; we take  $t^*$  as the unit of energy ( $t^* = 1$ ). The method of calculation is formulated for arbitrary values of  $s$  and  $S$  labels, but the results are presented for the spinless model ( $2s+1=1$  and  $2S+1=1$ ), and for the spin- $\frac{1}{2}$  model ( $2s+1=2$  and  $2S+1=2$ ). We consider only the homogeneous phase, where all quantities are translationally invariant.

### 2.1. Mapping on to the Falicov–Kimball atom

Infinite-coordination lattices can be solved by a mean-field-like procedure, because the self-energy of the conduction electrons is local (Metzner and Vollhardt 1989). That is, the Dyson's equation for the local d-electron Green's function  $G_{\text{loc}}^\sigma(z)$  on the lattice is

$$G_{\text{loc}}^\sigma(z) = \int \frac{\rho(\epsilon)}{z + \mu - \Sigma^\sigma(z) - \epsilon} d\epsilon, \quad (2)$$

where  $z$  is a complex variable and  $\Sigma^\sigma$  is the momentum-independent self energy. Hence, as noted by Brandt and Mielsch (1989), the lattice self-energy coincides with the self-energy of an atomic d state coupled to an f state by the same Coulomb interaction as on the lattice, and perturbed by an external time-dependent field  $\lambda^\sigma(\tau, \tau')$ . For an appropriate choice of the  $\lambda$  field, the functional dependence of  $\Sigma^\sigma$  on  $G^\sigma(z)$  and  $F^\eta(z)$ , the atomic propagators for d and f states, is exactly the same as in the lattice case. The lattice problem is thus reduced to finding the atomic self-energy functional for d electrons, and then setting  $G_{\text{loc}}^\sigma(z) = G^\sigma(z)$  and  $F_{\text{loc}}^\eta(z) = F^\eta(z)$  at each lattice site.

The FK atom can be solved by using the interaction representation, such that the time dependence of operators is defined by the atomic Hamiltonian

$$H_{\text{at}} = -\mu \sum_{\sigma} d_{\sigma}^{\dagger} d_{\sigma} + (E_{\text{f}} - \mu) \sum_{\eta} f_{\eta}^{\dagger} f_{\eta} + U \sum_{\sigma\eta} d_{\sigma}^{\dagger} d_{\sigma} f_{\eta}^{\dagger} f_{\eta}, \quad (3)$$

and the time dependence of the state vectors is governed by the evolution operator

$$S(\lambda) = T_{\tau} \exp \left( - \int_0^{\beta} d\tau \int_0^{\beta} d\tau' \sum_{\sigma} \lambda^{\sigma}(\tau, \tau') d_{\sigma}^{\dagger}(\tau) d_{\sigma}(\tau') \right). \quad (4)$$

The external field  $\lambda^{\sigma}(\tau, \tau')$  is assumed to be time translation invariant in (imaginary) time;

$$\lambda^{\sigma}(\tau, \tau') = T \sum_n \lambda_n^{\sigma} \exp[-i\omega_n(\tau - \tau')], \quad (5)$$

and hence can be expanded in a Fourier series in the fermionic Matsubara frequencies  $\omega_n = \pi(2n + 1)T$ , where we set  $k_{\text{B}} = 1$ . In the absence of an external magnetic field, the  $\lambda$  field is the same for all the  $\sigma$  components. The unperturbed atomic Hamiltonian (3) conserves the number of f and d electrons, while the time-dependent  $\lambda$  field gives rise to fluctuations in the d occupancy. In the equivalent lattice problem, the local d fluctuations are due to the d electron hopping.

The thermodynamics of the FK atom follows from the partition function

$$\mathcal{Z}_{\text{at}}(\lambda) = \text{Tr}_{\text{df}} [T_{\tau} \exp(-\beta H_{\text{at}}) S(\lambda)], \quad (6)$$

where the statistical sum runs over all possible atomic configurations, which is determined by the function  $\lambda^{\sigma}(\tau, \tau')$  for  $\tau, \tau' \in (0, \beta)$ . The specific feature of the atomic FK model is that the number of f electrons is a constant of motion and is either zero or one, while the d-electron number can assume any value between zero and  $2s + 1$ . Furthermore, the evolution operator  $S(\lambda)$  never transfers the state vectors out of the invariant ( $n_{\text{f}} = 0$  or  $n_{\text{f}} = 1$ ) Hilbert subspaces; so the matrix elements can be evaluated within each invariant subspace by replacing the operator  $\sum_{\eta} f_{\eta}^{\dagger} f_{\eta}$  by its eigenvalue 0 or 1. The trace in equation (6) can thus be performed separately for the  $n_{\text{f}} = 0$  and  $n_{\text{f}} = 1$  subspaces. Within the  $n_{\text{f}} = 0$  subspace, the operator dynamics are governed by a simplified non-interacting Hamiltonian

$$H_0 = -\mu \sum_{\sigma} d_{\sigma}^{\dagger} d_{\sigma}, \quad (7)$$

and we have  $d_{\sigma}^{\dagger}(\tau) = d_{\sigma}^{\dagger} \exp(-\mu\tau)$  and  $d_{\sigma}(\tau) = d_{\sigma} \exp(\mu\tau)$ , where  $d_{\sigma}^{\dagger} = d_{\sigma}^{\dagger}(0)$  and  $d_{\sigma} = d_{\sigma}(0)$  are the time-independent Schrödinger operators. The operator dynamics in the  $n_{\text{f}} = 1$  subspace are governed by the same Hamiltonian as in the  $n_{\text{f}} = 0$  sub-

space but with  $\mu$  replaced by  $\mu - U$ . The trace over f states in equation (6) can be performed, and we find that

$$\mathcal{Z}_{\text{at}}(\lambda) = \mathcal{Z}_0(\lambda, \mu) + (2S + 1) \exp[-\beta(E_f - \mu)] \mathcal{Z}_0(\lambda, \mu - U), \quad (8)$$

where

$$\mathcal{Z}_0(\lambda) = \prod_{\sigma} \mathcal{Z}_0^{\sigma}(\lambda^{\sigma}), \quad (9)$$

and

$$\mathcal{Z}_0^{\sigma}(\lambda^{\sigma}) = \text{Tr}_{\text{d}} [T_{\tau} \exp(-\beta H_0^{\sigma}) S(\lambda^{\sigma})]. \quad (10)$$

The factorization (9) holds because the time evolution due to  $H_0$  is such that the operators with different  $\sigma$  labels commute regardless of their time arguments, and the  $S$  matrix (4) does not change the  $\sigma$  label of a given state vector. Thus, the Hilbert space can be decomposed into invariant  $\sigma$  subspaces and the trace in equation (10) is over the non-degenerate  $d_{\sigma}$  states. In each of these subspaces, the operator dynamics are defined by  $H_0^{\sigma}$ , and the partition function  $\mathcal{Z}_0^{\sigma}(\lambda^{\sigma})$  describes the statistics of a  $d_{\sigma}$  electron subject to an arbitrary time-dependent  $\lambda^{\sigma}$  field.

In the presence of a magnetic field, the Hamiltonians (1) and (3) have to be supplemented with Zeeman terms

$$H_Z = g_{\text{d}} \mu_{\text{B}} H \sum_{\sigma} \sigma d_{\sigma}^{\dagger} d_{\sigma} + g_{\text{f}} \mu_{\text{B}} H \sum_{\eta} \eta f_{\eta}^{\dagger} f_{\eta}, \quad (11)$$

where  $g_{\text{d}}$  and  $g_{\text{f}}$  are the g factors of d and f electrons respectively. The solution of the model for  $H \neq 0$  is a straightforward generalization of the  $H = 0$  case, which is presented here (the main difference is that the effective chemical potentials now have a spin index dependence).

## 2.2. Generalized partition function for the Falicov–Kimball atom

The self-energy functional for the FK atom is calculated by using the equations of motion (EOMs) for the Green's function obtained by functional differentiation of the generalized partition function  $\mathcal{Z}_{\text{at}}(\lambda)$  (Kadanoff and Baym 1962). Equations (8)–(10) show that we can find  $\mathcal{Z}_{\text{at}}(\lambda)$  by solving a statistical problem for a single non-degenerate  $d_{\sigma}$  state coupled to the periodic  $\lambda^{\sigma}$ -field. Consider the contribution to  $\mathcal{Z}_0^{\sigma}(\lambda^{\sigma})$  due to the shift of the  $\lambda$  field from an initial configuration  $\lambda^{\sigma}(\tau, \tau')$  to the final configuration  $\lambda^{\sigma}(\tau, \tau') + \delta\lambda^{\sigma}(\tau, \tau')$ ;

$$\delta \mathcal{Z}_0^{\sigma} = \text{Tr}_{\text{d}} \langle T_{\tau} \exp(-\beta H_0^{\sigma}) \delta S(\lambda^{\sigma}) \rangle. \quad (12)$$

$\delta S(\lambda^{\sigma})$  is obtained from the usual rules of the calculus of variations;

$$\begin{aligned} \delta S(\lambda^{\sigma}) &= \delta \left[ \exp \left( - \int_0^{\beta} d\tau \int_0^{\beta} d\tau' \lambda^{\sigma}(\tau, \tau') d_{\sigma}^{\dagger}(\tau) d_{\sigma}(\tau') \right) \right] \\ &= -S(\lambda) \int_0^{\beta} d\tau \int_0^{\beta} d\tau' \delta\lambda^{\sigma}(\tau, \tau') d_{\sigma}^{\dagger}(\tau) d_{\sigma}(\tau'), \end{aligned} \quad (13)$$

and the time ordering is taken into account when this result is substituted into equation (12). Performing the substitution gives

$$\delta (\ln \mathcal{Z}_0^{\sigma}) = \int_0^{\beta} d\tau \int_0^{\beta} d\tau' \delta\lambda^{\sigma}(\tau, \tau') G_0^{\sigma}(\tau', \tau), \quad (14)$$

where

$$G_0^\sigma(\tau, \tau') = -\frac{1}{\mathcal{Z}_0} \text{Tr}_d [T_\tau \exp(-\beta H_0) d_\sigma(\tau) d_\sigma^\dagger(\tau') S(\lambda)] \quad (15)$$

is the d-electron Green's function restricted to configurations with no f electrons. The function multiplying the variation  $\delta\lambda^\sigma(\tau', \tau)$  is, by definition, the functional derivative of the partition function:

$$G_0^\sigma(\tau, \tau') = -\frac{\delta(\ln \mathcal{Z}_0^\sigma)}{\delta\lambda^\sigma(\tau', \tau)}. \quad (16)$$

Expressing  $G_0^\sigma(\tau, \tau')$  in equation (14) in terms of its Fourier components, that is

$$G_0^\sigma(\tau, \tau') = T \sum_n G_{0n}^\sigma \exp[-i\omega_n(\tau - \tau')], \quad (17)$$

and using equation (5) for  $\delta\lambda^\sigma(\tau, \tau')$  leads to the functional relation

$$\delta(\ln \mathcal{Z}_0^\sigma) = \sum_n G_{0n}^\sigma \delta\lambda_n^\sigma, \quad (18)$$

where  $G_{0n}^\sigma$  is now defined as a simple partial derivative:

$$G_{0n}^\sigma = -\frac{\partial(\ln \mathcal{Z}_0^\sigma)}{\partial\lambda_n^\sigma}. \quad (19)$$

An explicit expression for  $G_{0n}^\sigma(\lambda_n^\sigma)$  would allow us to obtain  $\mathcal{Z}_0^\sigma$  by solving the differential equation (19). The functions  $G_{0n}^\sigma$  and  $\lambda_n^\sigma$ , or  $G_0^\sigma(\tau, \tau')$  and  $\lambda^\sigma(\tau', \tau)$ , can be considered as matrix elements of integral operators  $G_0^\sigma$  and  $\lambda^\sigma$ , and equations (14) and (19) can be written in the operator form as

$$\delta(\ln \mathcal{Z}_0^\sigma) = \text{Tr}(G_0^\sigma \delta\lambda^\sigma), \quad (20)$$

where the trace denotes an integration over time if we are using non-diagonal matrices in the  $\tau$  representation, or a Matsubara summation, if we are using diagonal matrices in frequency space.

Our next step is to find an explicit expression for  $G_{0n}^\sigma$  and to solve equation (19) to find  $\mathcal{Z}_0^\sigma$ . The Green's function in equations (15) and (19) are obtained by the EOM. Consider first the case  $\tau > \tau'$ . To compute  $(\partial/\partial\tau)G_0^\sigma(\tau, \tau')$ , we must first compute the derivative of  $[T_\tau d_\sigma(\tau) d_\sigma^\dagger(\tau') S(\lambda^\sigma)]$  with respect to  $\tau$ . It is important to note that the differential operator does not commute with the time-ordering operator; so one must proceed carefully. Note that when we take a derivative with respect to  $S(\lambda^\sigma)$ , it will bring down terms from the exponent such as  $d_\sigma^\dagger(\tau) d_\sigma(\tau_2)$  or  $d_\sigma^\dagger(\tau_1) d_\sigma(\tau)$ , and the latter terms will not contribute when multiplied by  $d_\sigma(\tau)$ : that is the time ordering with respect to  $\tau_1$  is the only important variable to consider when taking the derivative. So we write the full-time ordered product in two pieces:

$$T_\tau d_\sigma(\tau) d_\sigma^\dagger(\tau') S(\lambda^\sigma) = [T_\tau \bar{S}(\lambda^\sigma)] d_\sigma(\tau) [T_\tau d_\sigma^\dagger(\tau') \bar{\bar{S}}(\lambda^\sigma)], \quad (21)$$

with

$$\begin{aligned} \bar{S}(\lambda^\sigma) &= \exp\left(-\int_\tau^\beta d\tau_1 \int_0^\beta d\tau_2 \lambda^\sigma(\tau_1, \tau_2) d_\sigma^\dagger(\tau_1) d_\sigma(\tau_2)\right), \\ \bar{\bar{S}}(\lambda^\sigma) &= \exp\left(-\int_0^\tau d\tau_1 \int_0^\beta d\tau_2 \lambda^\sigma(\tau_1, \tau_2) d_\sigma^\dagger(\tau_1) d_\sigma(\tau_2)\right). \end{aligned} \quad (22)$$

Now the derivative can be computed directly to yield

$$\frac{\partial}{\partial \tau} \{T_\tau[d_\sigma(\tau)d_\sigma^\dagger(\tau')S(\lambda^\sigma)]\} = T_\tau \left[ \left( \mu d_\sigma(\tau) - \int_0^\beta d\tau_2 \lambda^\sigma(\tau, \tau_2) d_\sigma(\tau_2) \right) d_\sigma^\dagger(\tau') S(\lambda^\sigma) \right], \quad (23)$$

where we employed identities such as  $d_\sigma(\tau_2)d_\sigma^\dagger(\tau)d_\sigma(\tau) = d_\sigma(\tau_2)$  for  $\tau_2 > \tau$  (which can be easily derived from the fact that the time dependence of the operators involves only an exponential factor and the anticommutator of the Fermionic operators is 1). Since the form for the Green's function is different for  $\tau < \tau'$ , one must repeat the derivation there (with the same result). Hence the Green's function satisfies the following EOM:

$$\left( -\frac{\partial}{\partial \tau} + \mu \right) G_0^\sigma(\tau, \tau') - \int_0^\beta d\tau'' \lambda^\sigma(\tau, \tau'') G_0^\sigma(\tau'', \tau') = \delta(\tau - \tau'), \quad (24)$$

where the  $\delta$  function arises from the discontinuity in  $G_0$  at  $\tau = \tau'$ . This EOM can also be written as

$$\int_0^\beta d\tau'' [G_0^\sigma]^{-1}(\tau, \tau'') G_0^\sigma(\tau'', \tau') = \delta(\tau - \tau'), \quad (25)$$

or, in matrix representation,

$$[G_0^\sigma]^{-1} G_0^\sigma = \mathbf{1}, \quad (26)$$

where  $[G_0^\sigma]^{-1}$  is the non-diagonal integral operator defined by its matrix elements

$$[G_0^\sigma]^{-1}(\tau, \tau') = \left( -\frac{\partial}{\partial \tau} + \mu \right) \delta(\tau - \tau') - \lambda^\sigma(\tau, \tau'), \quad (27)$$

and the unit operator  $\mathbf{1}$  has the matrix elements  $\delta(\tau - \tau')$ . The Fourier transform of equation (27) gives the matrix elements of  $[G_0^\sigma]^{-1}$  as

$$[G_0^\sigma]^{-1}(i\omega_n) = i\omega_n + \mu - \lambda_n^\sigma, \quad (28)$$

and of its inverse  $G_0^\sigma$  as

$$G_0^\sigma(i\omega_n) = \frac{1}{i\omega_n + \mu - \lambda_n^\sigma}. \quad (29)$$

The diagonal forms of  $[G_0^\sigma]^{-1}$  and  $G_0^\sigma$  in Fourier space are the consequence of the time translation invariance of  $\lambda^\sigma(\tau, \tau')$ . Using equations (19) and (29) we obtain the differential equation (Kadanoff and Baym 1962)

$$\frac{1}{i\omega_n + \mu - \lambda_n^\sigma} = -\frac{\partial(\ln \mathcal{Z}_0^\sigma)}{\partial \lambda_n^\sigma}, \quad (30)$$

which has to be solved together with the initial ( $\lambda = 0$ ) boundary condition,

$$\mathcal{Z}_0^\sigma(0, \mu) = 1 + \exp(\beta\mu). \quad (31)$$

The partition function for the simplified atomic problem is thus obtained as (Brandt and Mielsch 1989)

$$\mathcal{Z}_0^\sigma(\lambda^\sigma, \mu) = 2 \exp(\beta\mu/2) \prod_n \frac{i\omega_n + \mu - \lambda_n^\sigma}{i\omega_n} \quad (32)$$

and the complete partition function for the FK atom can be written as

$$\mathcal{Z}_{\text{at}}(\lambda) = \prod_{\sigma} \mathcal{Z}_0^{\sigma}(\lambda^{\sigma}, \mu) + (2S + 1) \exp[-\beta(E_f - \mu)] \prod_{\sigma} \mathcal{Z}_0^{\sigma}(\lambda^{\sigma}, \mu - U). \quad (33)$$

### 2.3. Dynamics of the atomic $d$ state

The renormalized  $d$ -electron propagator can be obtained, in complete analogy to equations (12)–(16), as a functional derivative of  $\mathcal{Z}_{\text{at}}$  with respect to the external field:

$$G^{\sigma}(\tau, \tau') = -\frac{\delta(\ln \mathcal{Z}_{\text{at}})}{\delta \lambda^{\sigma}(\tau', \tau)} \quad (34)$$

such that

$$G_{\text{at}}^{\sigma}(\tau, \tau') = -\frac{1}{\mathcal{Z}_{\text{at}}} \text{Tr}_{\text{df}} \langle T_{\tau} \exp(-\beta H_{\text{at}}) d_{\sigma}(\tau) d_{\sigma}^{\dagger}(\tau') S(\lambda^{\sigma}) \rangle. \quad (35)$$

The difference with respect to the  $n_f = 0$  case is that the trace extends now over the  $d$  and  $f$  states, including the  $\sigma$  and  $\eta$  labels, and the statistical operator is the full atomic Hamiltonian  $H_{\text{at}}$  rather than  $H_0^{\sigma}$ . On the imaginary frequency axis we still have

$$G_n^{\sigma} = -\frac{\partial(\ln \mathcal{Z}_{\text{at}})}{\partial \lambda_n^{\sigma}}, \quad (36)$$

which gives, using equations (32) and (33), the result

$$G_n^{\sigma} = \frac{w_0}{[G_{0n}^{\sigma}]^{-1}} + \frac{w_1}{[G_{0n}^{\sigma}]^{-1} - U}, \quad (37)$$

where  $w_0 = \mathcal{Z}_0/\mathcal{Z}_{\text{at}}$  and  $w_1 = 1 - w_0$ . The weight  $w_1$  gives the  $f$  occupation number (Brandt and Mielsch 1989).

On the other hand, starting from the definition of the Green's function in equation (35), and making the usual Feynman–Dyson expansion with  $U$  as the expansion parameter, we obtain the standard Feynman diagrams, in terms of the unperturbed propagators  $G_{0n}^{\sigma}$ . The self-energy function  $\Sigma_n^{\sigma}$  of the FK atom on the imaginary frequency axis is defined by the Dyson equation

$$\Sigma_n^{\sigma} = [G_{0n}^{\sigma}]^{-1} - [G_n^{\sigma}]^{-1}. \quad (38)$$

Eliminating  $G_{0n}^{\sigma}$ , and hence the  $\lambda$  field, from equations (37) and (38) yields

$$\Sigma_n^{\sigma} = \frac{w_1 U}{1 - (U - \Sigma_n^{\sigma}) G_{n\sigma}}, \quad (39)$$

which can also be written in the form given by Brandt and Mielsch (1989):

$$\Sigma_n^{\sigma} = \frac{1}{2} \left\{ U - \frac{1}{G_{n\sigma}} \pm \left[ \left( \frac{1}{G_{n\sigma}} - U \right)^2 + 4w_1 \frac{U}{G_{n\sigma}} \right]^{1/2} \right\}, \quad (40)$$

where the sign of the square root is chosen to maintain the proper analyticity properties of  $\Sigma$ .

To clarify the physical meaning of the self-energy (39) we now perform the calculations directly for the original lattice model (1) using the EOM. Since we also have to consider the Green's function of higher order in the creation and

annihilation operators, it is convenient to introduce the compact Zubarev notation, where the Fourier-transformed quantities are written as,

$$\langle\langle A, B \rangle\rangle_n = \int_0^\beta d(\tau - \tau') \exp[i\omega_n(\tau - \tau')] \langle T_\tau A(\tau) B(\tau') \rangle, \quad (41)$$

and  $\langle \dots \rangle$  denotes the thermal averaging over all the states on the lattice:

$$\langle \hat{O} \rangle = \frac{1}{Z} \text{Tr}_{\text{df}} [\exp(-\beta H) \hat{O}]. \quad (42)$$

The Fourier transform of the lattice Green's function at site  $i$  is now written as  $G_{n\sigma} = \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n$ , and the EOM is

$$(\mu + i\omega_n) \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n = 1 - \sum_j t_{ij} \langle\langle d_{j\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n + U \sum_\eta \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, d_{i\sigma}^\dagger \rangle\rangle_n. \quad (43)$$

Using the EOM for the higher-order Green's function on the right-hand side of equation (43), and considering also the time derivative with respect to the second time variable  $\tau'$ , we obtain

$$\begin{aligned} (\mu + i\omega_n) \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, d_{i\sigma}^\dagger \rangle\rangle_n &= \langle f_{i\eta}^\dagger f_{i\eta} \rangle - \sum_j t_{ij} \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, d_{j\sigma}^\dagger \rangle\rangle_n \\ &+ U \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, f_{i\eta}^\dagger f_{i\eta} d_{i\sigma}^\dagger \rangle\rangle_n. \end{aligned} \quad (44)$$

Here, we used the fact that a given site  $i$  cannot be occupied by more than one f electron, which means that the same spin index  $\eta$  appears for the f-electron operators at both times  $\tau$  and  $\tau'$ . As the f occupation per site is conserved, we also have the relation

$$\langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, f_{i\eta}^\dagger f_{i\eta} d_{i\sigma}^\dagger \rangle\rangle_n = \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, d_{i\sigma}^\dagger \rangle\rangle_n. \quad (45)$$

Defining the local (site-diagonal) self-energy  $\Sigma_n^\sigma$  as

$$\Sigma_n^\sigma \langle\langle d_{i\sigma}, d_{j\sigma}^\dagger \rangle\rangle_n = U \sum_\eta \langle\langle d_{i\sigma} f_{i\eta}^\dagger f_{i\eta}, d_{j\sigma}^\dagger \rangle\rangle_n \quad (46)$$

we obtain, from equation (44),

$$\Sigma_n^\sigma \left( (\mu + i\omega_n) \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n + \sum_j t_{ij} \langle\langle d_{i\sigma}, d_{j\sigma}^\dagger \rangle\rangle_n \right) = U \sum_\eta \langle f_{i\eta}^\dagger f_{i\eta} \rangle + U \Sigma_n^\sigma \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n. \quad (47)$$

Denoting the total average f occupation per site  $i$  by  $w_1 = \sum_\eta \langle f_{i\eta}^\dagger f_{i\eta} \rangle$ , and using

$$(\mu + i\omega_n) \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n + \sum_j t_{ij} \langle\langle d_{i\sigma}, d_{j\sigma}^\dagger \rangle\rangle_n = 1 + \Sigma_n^\sigma \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n, \quad (48)$$

which follows from equation (43), we find that

$$\Sigma_n^\sigma [1 - (U - \Sigma_n^\sigma) \langle\langle d_{i\sigma}, d_{i\sigma}^\dagger \rangle\rangle_n] = U w_1, \quad (49)$$

which is equivalent to equation (39) and which we recognize as the standard Hubbard III (coherent-phase approximation (CPA)) self-consistency equation for the self-energy. The fact that the exact d-electron self-energy is given by the CPA, which becomes exact in the limit of infinite dimensions for disordered systems, has a

simple physical interpretation. As the f-electron number per site is conserved, the d electrons ‘see’ an effective disordered alloy potential because at a certain site  $i$  there is either the on-site potential 0, if the site is not occupied by an f electron, with probability  $w_0$ , or there is the on-site potential  $U$ , if the site is occupied by an f electron, with probability  $w_1 = 1 - w_0$ . However, the self-energy functional depends explicitly on  $w_1$ , which is not known unless one calculates the full partition function.

### § 3. FORMALISM FOR THE f-ELECTRON GREEN’S FUNCTION

The atomic f propagator  $F_\eta(\tau)$  is defined in the interaction representation for  $\tau > 0$  as (Brandt and Urbanek 1992)

$$F_\eta(\tau) = \frac{-\text{Tr}_{\text{df}} [\exp(-\beta H_{\text{at}}) S(\lambda) f_\eta(\tau) f_\eta^\dagger(0)]}{\mathcal{Z}_{\text{at}}(\lambda)}, \quad (50)$$

where the trace is taken over the atomic d and f states, including the spin labels, and  $H_{\text{at}}$  is the full atomic Hamiltonian defined by equation (3). The time evolution of the f and d operators is now defined as  $f_\eta(\tau) = \exp(\tau H_{\text{at}}) f_\eta \exp(-\tau H_{\text{at}})$ , and  $d_\sigma^\dagger(\tau) = \exp(\tau H_{\text{at}}) d_\sigma^\dagger \exp(-\tau H_{\text{at}})$ , which leads to the EOM,

$$\frac{d}{d\tau} [d_\sigma^\dagger(\tau)] = \left( -\mu + U \sum_\eta f_\eta^\dagger(\tau) f_\eta(\tau) \right) d_\sigma^\dagger(\tau), \quad (51)$$

$$\frac{d}{d\tau} [f_\eta(\tau)] = \left( -(E_f - \mu) - U \sum_\sigma d_\sigma^\dagger(\tau) d_\sigma(\tau) \right) f_\eta(\tau). \quad (52)$$

The integral representation of equations (51) and (52) is

$$d_\sigma^\dagger(\tau) = \exp(-\mu\tau) T_\tau \exp \left( U \sum_\eta \int_0^\tau d\tau'' f_\eta^\dagger(\tau'') f_\eta(\tau'') \right) d_\sigma^\dagger, \quad (53)$$

$$f_\eta(\tau) = \exp[-(E_f - \mu)\tau] T_\tau \exp \left( -U \sum_\sigma \int_0^\tau d\tau'' d_\sigma^\dagger(\tau'') d_\sigma(\tau'') \right) f_\eta, \quad (54)$$

which can be written as

$$f_\eta(\tau) = \exp[-(E_f - \mu)\tau] S'(\tau) f_\eta. \quad (55)$$

Here  $S'(\tau)$  is the time-ordered exponential:

$$S'(\tau) = T_\tau \exp \left( - \sum_\sigma \int_0^\beta d\tau' \int_0^\beta d\tau'' \chi_\tau(\tau', \tau'') d_\sigma^\dagger(\tau') d_\sigma(\tau'') \right), \quad (56)$$

and

$$\chi_\tau(\tau', \tau'') = U \Theta(\tau - \tau') \delta(\tau' - \tau''), \quad (57)$$

with  $\Theta(x)$  the unit step function:  $\Theta(x > 0) = 1$  and  $\Theta(x < 0) = 0$ . Equations (53) and (55) lead to the expression

$$F_\eta(\tau) = \frac{-\exp[-(E_f - \mu)\tau] T_\tau \text{Tr}_{\text{df}} [\exp(-\beta H_{\text{at}}) S(\lambda) S'(\chi_\tau) f_\eta f_\eta^\dagger]}{\mathcal{Z}_{\text{at}}(\lambda)}, \quad (58)$$

which shows that the operator  $f_\eta f_\eta^\dagger(0)$  and the constraint  $n_f = \sum_\eta f_\eta f_\eta^\dagger \leq 1$  eliminates all the  $n_f = 1$  states from the trace in equation (58). In addition, all the intermediate states of the system, obtained by applying  $SS'$  to an initial state in the  $n_f = 0$  subspace, remain in the same subspace. This is because the  $f$  operators in  $S$  and  $S'$  always appear as equal-time pairs,  $f_\eta^\dagger(\tau')f_\eta(\tau')$ , and just count the  $f$  electrons at time  $\tau'$ . Thus, the statistical problem for  $F_\eta(\tau)$  is restricted to a constant  $n_f$  subspace and we can replace the operator expression  $\sum_\eta = f_\eta^\dagger f_\eta$  in  $H_{\text{at}}$  and in  $SS'$  by its eigenvalue 0 or 1. We use  $n_f = 0$  for  $\tau > 0$  propagation and  $n_f = 1$  for  $\tau < 0$  propagation. The  $f$ -electron Green's function for  $\tau \geq 0$  becomes

$$F_\eta(\tau) = \frac{-\exp[-(E_f - \mu)\tau] \text{Tr}_d[\exp(-\beta H_0) S(\lambda) S'(\tau)]}{\mathcal{Z}_{\text{at}}(\lambda)}, \quad (59)$$

with the statistical operator defined once again by  $H_0$  in equation (7) and the statistical averaging performed with respect to all possible states of a  $d$  electron perturbed by the  $\lambda$  field and by the additional time-dependent field  $\chi_\tau$ .

In the interaction representation defined by  $H_0$ , the time dependence of the annihilation and creation operators is again  $d_\sigma(\tau) = \exp(\mu\tau) d_\sigma$  and  $d_\sigma^\dagger(\tau) = \exp(-\mu\tau) d_\sigma^\dagger$ , and the time ordering becomes trivial. Thus, the product of two  $T$ -ordered exponentials in equation (59) can be written as a single  $T$ -ordered exponential:

$$S(\tilde{\lambda}_\sigma) = T_\tau \exp\left(-\sum_\sigma \int_0^\beta d\tau' \int_0^\beta d\tau'' \tilde{\lambda}_\sigma(\tau', \tau'') d_\sigma^\dagger(\tau') d_\sigma(\tau'')\right), \quad (60)$$

where

$$\tilde{\lambda}_\sigma(\tau', \tau'') = \lambda^\sigma(\tau', \tau'') - \chi_\tau(\tau', \tau''), \quad (61)$$

which also depends on the external time  $\tau$ . The  $f$ -electron Green's function becomes

$$F_\eta(\tau) = \frac{-\exp[-(E_f - \mu)\tau] T_\tau \text{Tr}_d[\exp(-\beta H_0) S(\tilde{\lambda})]}{\mathcal{Z}_{\text{at}}(\lambda)}, \quad (62)$$

and the problem is reduced to the evaluation of the statistical sum of a single atomic  $d$  level coupled to a time-dependent  $\tilde{\lambda}$  field.

### 3.1. *Effective partition function for the $f$ -electron problem*

The effective partition function required for the  $f$  propagator can be written in the factorized form,

$$\mathcal{Z}_0(\tilde{\lambda}) = T_\tau \text{Tr}_d[\exp(-\beta H_0) S(\tilde{\lambda})] = \prod_\sigma \mathcal{Z}_0^\sigma(\tilde{\lambda}_\sigma). \quad (63)$$

The factorization (63) holds because the time evolution due to  $H_0$  is such that the annihilation and creation operators with different spin labels commute, regardless of their time arguments, and the exponential operator  $S(\tilde{\lambda})$  can be factorized.

The time-translation-invariant component of the  $\tilde{\lambda}$  field is determined by mapping the FK atom on to the FK lattice, while the additional  $\chi_\tau$  component is defined by the Coulomb interaction between the  $f$  and  $d$  electrons during the time interval  $(0, \tau)$ . The presence of this additional time-dependent field can be understood as follows. In the FK atom the operator dynamics are defined by  $H_{\text{at}}$  and the  $d$  occupancy of the state vector is time dependent because of the  $\lambda$  field. The interaction

between f and d electrons gives rise to fluctuations in the f-level position and makes the potential energy of the system time dependent. In the effective d-electron system described by equation (62), the changes in interaction energy due to the fluctuating f–d interaction energy are represented by the  $\chi_\tau$  field. In other words, the f electrons with an infinite lifetime acquire dynamics because of the coupling to the d-electron fluctuations. In this respect, the FK problem is similar to the X-ray edge problem (Si *et al.* 1992). However, while the X-ray problem is formulated as a single-site problem, in the FK atom the self-consistently determined  $\lambda$  field keeps track of all other f sites in the lattice. At high temperatures, where the coherent scattering of conduction electrons on the f ions could be neglected, the single-site model (where there is no self-consistency to determine the  $\lambda$  field) might be representative of the lattice (Si *et al.* 1992) but at low temperatures, where coherence develops, the single-site description and the X-ray analogy might not be appropriate, and the lattice effects due to the  $\lambda$  field have to be taken into account.

The partition function  $Z_0(\tilde{\lambda})$  cannot be calculated with the same procedure as for  $Z_0(\lambda)$  because the  $\tilde{\lambda}$  field is no longer a function of  $\tau - \tau'$ . Thus, the effective propagator associated with  $Z_0(\tilde{\lambda})$  cannot be diagonalized by a Fourier transformation, and a simple differential equation for  $Z_0(\tilde{\lambda})$  cannot be derived. Nonetheless, we use the functional differentiation of  $Z_0(\tilde{\lambda})$  with respect to  $\tilde{\lambda}$  to generate an effective Green's function

$$g_\sigma(\tau', \tau'') = -\frac{\delta\{\ln [Z_0^\sigma(\tilde{\lambda}_\sigma)]\}}{\delta\tilde{\lambda}_\sigma(\tau'', \tau')} \quad (64)$$

such that

$$g_\sigma(\tau', \tau'') = -\frac{1}{Z_0^\sigma(\tilde{\lambda}_\sigma)} \text{Tr}_d \langle T_\tau \exp(-\beta H_0^\sigma) d_\sigma(\tau') d_\sigma^\dagger(\tau'') S(\tilde{\lambda}_\sigma) \rangle. \quad (65)$$

Similarly, we introduce an auxiliary Green's function defined for a d level driven by the  $\chi_\tau$  field only:

$$g_{0\sigma}(\tau', \tau'') = -\frac{\delta\{\ln [Z_0^\sigma(\chi_\tau)]\}}{\delta\chi_\tau(\tau'', \tau')}, \quad (66)$$

where

$$Z_0^\sigma(\chi_\tau) = T_\tau \text{Tr}_d [\exp(-\beta H_0) S(\chi_\tau)], \quad (67)$$

and

$$g_{0\sigma}(\tau', \tau'') = -\frac{1}{Z_0^\sigma(\chi_\tau)} \text{Tr}_d \langle T_\tau \exp(-\beta H_0^\sigma) d_\sigma(\tau') d_\sigma^\dagger(\tau'') S(\chi_\tau) \rangle. \quad (68)$$

The Green's functions  $g_\sigma$  and  $g_{0\sigma}$  depend explicitly on times  $\tau'$  and  $\tau''$ , and implicitly on  $\tau$ .

The evaluation of  $Z_0^\sigma(\chi_\tau)$  and  $g_{0\sigma}$  is straightforward because the evolution operator  $S(\chi_\tau)$  does not change the number of d electrons. The Hilbert space for d states, in the absence of the  $\lambda$  field, consists of only two states ( $n_d = 0$  and  $n_d = 1$ ), and the result for the partition function is simply

$$Z_0^\sigma(\chi_\tau) = 1 + \exp(\beta\mu - U\tau) \quad (69)$$

and

$$\mathcal{Z}_0(\chi_\tau) = \prod_\sigma \mathcal{Z}_0^\sigma(\chi_\tau). \tag{70}$$

The time-ordered product in equation (68) has to be treated with some care, because the functional form of the  $\chi_\tau$  field is not the same in all the parts of the  $(\tau', \tau'')$  plane (see equation (57)). Eventually, we obtain the following expressions for  $g_{0\sigma}(\tau', \tau'')$  (Brandt and Urbanek 1992):

$$g_{0a^+} = (\xi_0 - 1) \exp [(\mu - U)\tau'] \exp [-(\mu - U)\tau''], \quad \text{for } \tau'' < \tau' < \tau, \tag{71}$$

$$g_{0a^-} = \xi_0 \exp [(\mu - U)\tau'] \exp [-(\mu - U)\tau''], \quad \text{for } \tau' < \tau'' < \tau, \tag{72}$$

$$g_{0b^+} = (\xi_0 - 1) \exp (-U\tau) \exp (\mu\tau') \exp [-(\mu - U)\tau''], \quad \text{for } \tau'' < \tau < \tau', \tag{73}$$

$$g_{0b^-} = \xi_0 \exp (U\tau) \exp [(\mu - U)\tau'] \exp (-\mu\tau''), \quad \text{for } \tau' < \tau < \tau'', \tag{74}$$

$$g_{0c^+} = (\xi_0 - 1) \exp (\mu\tau') \exp (-\mu\tau''), \quad \text{for } \tau < \tau'' < \tau', \tag{75}$$

$$g_{0c^-} = \xi_0 \exp (\mu\tau') \exp (-\mu\tau''), \quad \text{for } \tau < \tau' < \tau'', \tag{76}$$

where the + and - in the subscript relates to evaluating  $g_0(\tau', \tau'')$  above and below the line  $\tau' = \tau''$  respectively and the letters a, b, or c refer to a different region on the  $0-\beta$  square as depicted in figure 1. Here the symbol  $\xi_0 = 1/[1 + \exp (U\tau - \beta\mu)]$ . The function  $g_{0\sigma}(\tau', \tau'')$  is implicitly time dependent, because its functional form depends on the relative magnitude of  $\tau$  with respect to  $\tau'$  and  $\tau''$ .

To proceed, we consider the periodic  $\lambda$  field as an additional perturbation to the statistical problem defined by  $\mathcal{Z}_0(\chi_\tau)$ . Thus, writing the full  $S$  matrix in equation (56) in the factorized form  $S(\tilde{\lambda}) = S(\lambda)S(\chi_\tau)$ , we find that the Green's functions obtained by the functional derivatives  $\delta\{\ln [\mathcal{Z}(\tilde{\lambda})]\}/\delta\tilde{\lambda}(\tau'', \tau')$  and  $\delta\{\ln [\mathcal{Z}_0(\chi_\tau)]\}/\delta\chi_\tau(\tau'', \tau')$  are related by a Dyson equation. In the operator form, this can be written as

$$g_\sigma = g_{0\sigma} - g_{0\sigma}\lambda^\sigma g_\sigma \tag{77}$$

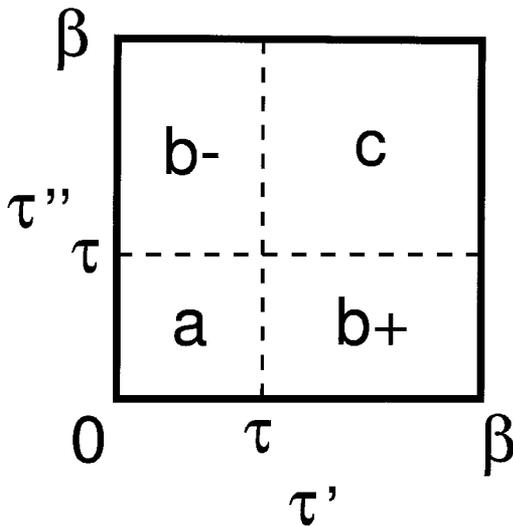


Figure 1. Different regions of the integration range for  $\tau'$  and  $\tau''$ .

or, equivalently,

$$g_\sigma^{-1} = g_{0\sigma}^{-1} - \lambda^\sigma = g_{0\sigma}^{-1}(1 - g_{0\sigma} \lambda^\sigma), \tag{78}$$

where  $g_\sigma$  and  $g_{0\sigma}$  are non-diagonal integral operators both in the time and in the frequency representation, while  $\lambda^\sigma$  is diagonal in the frequency representation. Next, we show that the partition function can be written as

$$\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma) = \det |g_\sigma^{-1}|. \tag{79}$$

This holds, because the functional derivatives  $\delta\{\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]\}/\delta\tilde{\lambda}^\sigma(\tau', \tau)$  and  $\delta\{\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]\}/\delta\lambda^\sigma(\tau', \tau)$  define the same Green's function  $g(\tau, \tau')$ , so that we can write

$$\begin{aligned} \delta\{\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]\} &= - \int_0^\beta d\tau \int_0^\beta d\tau' g_\sigma(\tau, \tau') \delta\lambda^\sigma(\tau', \tau) \\ &= - \int_0^\beta d\tau \int_0^\beta d\tau' g_\sigma(\tau, \tau') \delta\tilde{\lambda}^\sigma(\tau', \tau) \end{aligned} \tag{80}$$

where  $g_\sigma(\tau, \tau')$  is given by equation (65). In other words, the variation in  $\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]$  is not changed if  $\lambda^\sigma(\tau', \tau)$  is shifted with respect to some arbitrary but fixed surface  $[g_{0\sigma}]^{-1}(\tau', \tau)$ . Using equation (78) we write  $\delta\tilde{\lambda}^\sigma = \delta(\lambda^\sigma - g_{0\sigma}^{-1}) = -\delta g_\sigma^{-1}$  and obtain

$$\begin{aligned} \delta\{\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]\} &= - \int_0^\beta d\tau \int_0^\beta d\tau' g_\sigma(\tau, \tau') \delta(\lambda^\sigma - g_{0\sigma}^{-1})(\tau', \tau) \\ &= \int_0^\beta d\tau (g_\sigma \delta g_\sigma^{-1})(\tau, \tau), \end{aligned} \tag{81}$$

where in the last equation we arrived at the diagonal matrix elements of  $g_\sigma \delta g_\sigma^{-1}$  by carrying out the matrix multiplication of  $g_\sigma(\tau, \tau')$  and  $\delta g_\sigma^{-1}(\tau', \tau)$ . Since  $g_\sigma$  is the inverse of  $g_\sigma^{-1}$ , equation (81) can be written as

$$\delta\{\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)]\} = \int_0^\beta d\tau \delta[\ln (g_\sigma^{-1})(\tau, \tau)], \tag{82}$$

which shows that  $\delta(\ln \mathcal{Z}_0^\sigma)$  follows from the variation in  $\text{Tr}(\ln [g_\sigma^{-1}])$ . Thus,

$$\ln [\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma)] = \text{Tr}(\ln [g_\sigma^{-1}]), \tag{83}$$

and the matrix identity  $\text{Tr}(\ln A) = \ln(\det A)$  leads to equation (79). The Dyson equation (78) then yields the effective partition function as a continous determinant:

$$\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma) = \det |g_{0\sigma}^{-1}| \det |1 - g_{0\sigma} \lambda^\sigma|, \tag{84}$$

which can be written as

$$\mathcal{Z}_0^\sigma(\tilde{\lambda}^\sigma) = \mathcal{Z}_0^\sigma(\chi_\tau) \det |1 - g_{0\sigma} \lambda^\sigma| = \mathcal{Z}_0^\sigma(\chi_\tau) \det D^\sigma. \tag{85}$$

Here, we introduced the integral operator  $D^\sigma$ , which is defined in the  $\tau$  representation by its matrix elements:

$$D_{\tau_1, \tau_2}^\sigma = \delta(\tau_1 - \tau_2) - \int_0^\beta d\tau'' g_{0\sigma}(\tau_1, \tau'') \lambda^\sigma(\tau'', \tau_2). \tag{86}$$

The Fourier transform of equation (86) defines the integral operator  $D^\sigma$  in frequency space where its matrix elements form a discrete set. The time translation invariance of the  $\lambda$  field leads to the expression,

$$D_{pq}^\sigma = \delta_{pq} - \frac{\lambda_q^\sigma}{\beta} \int_0^\beta d\tau' \exp(i\omega_p \tau') \int_0^\beta d\tau'' g_{0\sigma}(\tau', \tau'') \exp(-i\omega_q \tau''), \quad (87)$$

where the integrals

$$M_{pq}^\sigma = \int_0^\beta d\tau' \exp(i\omega_p \tau') \int_0^\beta d\tau'' g_{0\sigma}(\tau', \tau'') \exp(-i\omega_q \tau'') \quad (88)$$

are given in Appendix A. The implicit dependence of  $g_{0\sigma}(\tau', \tau'')$  on the external time makes  $M_{pq}^\sigma$  explicitly  $\tau$  dependent. Since the determinant of an operator is the same in any basis, we can calculate  $\mathcal{Z}_0^\sigma(\tilde{\lambda})$  in the Matsubara representation using the discrete matrix elements (87).

The final form for the f-electron Green's function is thus (Brandt and Urbanek 1992)

$$F_\eta(\tau) = - \frac{\exp[-(E_f - U)\tau] \prod_\sigma [\mathcal{Z}_0^\sigma(\chi_\tau) \det D_{pq}^\sigma]}{\mathcal{Z}_{\text{at}}(\lambda)}. \quad (89)$$

It is useful to examine the expression in equation (89) for the limits  $\tau \rightarrow 0^+$  and  $\tau \rightarrow \beta^-$  for the spinless case. In the former case we have  $g_0(i\omega_p, i\omega_q) = \delta_{pq}/(i\omega_p + \mu)$  and

$$F(\tau = 0^+) = - \frac{2 \exp(\beta\mu/2) \prod_n (i\omega_n + \mu - \lambda_n)/(i\omega_n)}{\mathcal{Z}_{\text{at}}(\lambda)} = -w_0, \quad (90)$$

and in the latter case we have  $g_0(i\omega_p, i\omega_q) = \delta_{pq}/(i\omega_p + \mu - U)$  and

$$\begin{aligned} F(\tau = \beta^-) &= - \frac{\exp[-\beta(E_f - \mu)] 2 \exp[\beta(\mu - U)/2] \prod_n (i\omega_n + \mu - \lambda_n - U)/(i\omega_n)}{\mathcal{Z}_{\text{at}}(\lambda)} = -w_1, \end{aligned} \quad (91)$$

as we must have by directly evaluating the Green's function.

#### §4. FORMALISM FOR THE SPONTANEOUS HYBRIDIZATION

Recent work by Portengen *et al.* (1996a,b) proposed that the FK model may have a ground state that has a non-zero average for a spontaneous hybridization  $\langle df^\dagger \rangle$ . Such a state would imply that the FK fixed point is unstable to the periodic Anderson model fixed point. This instability is not allowed at any finite temperature because the conservation of the local f-electron number implies the existence of a local gauge symmetry which cannot be broken at finite temperature owing to Elitzur's (1975) theorem (see also Subrahmanya and Barma (1988)). This is the same conclusion arrived at from perturbation theory (Czycholl 1999) and exact diagonalization (Farkasovský, 1997, 1999). Here we shall show how to test these ideas exactly in the infinite-dimensional limit. (A mapping of the FK model with a Lorentzian density of states on to the X-ray edge problem shows that the hybridization susceptibility can diverge at  $T = 0$  (Si *et al.* 1992), but it is not clear that this behaviour will survive when one examines a conventional lattice.)

The static susceptibility for spontaneous hybridization satisfies

$$\chi_{\text{hyb}} = - \int_0^\beta d\tau \left( \frac{\text{Tr}_{\text{df}} \langle T_\tau \exp(-\beta H_{\text{at}}) S(\lambda) f(\tau) d^\dagger(\tau) d(0) f^\dagger(0) \rangle}{\mathcal{Z}_{\text{at}}(\lambda)} + G(\tau) F(\tau) \right), \quad (92)$$

for the spinless FK model. The integrand can be determined by simply taking the functional derivative of the f-electron Green's function with respect to  $\lambda(\tau, 0)$ :

$$\begin{aligned} \chi_{\text{hyb}} &= - \int_0^\beta d\tau \left( \frac{\delta F(\tau)}{\delta \lambda(\tau, 0)} + G(\tau) F(\tau) \right) \\ &= \int_0^\beta d\tau \frac{\exp[-(E_f - \mu)\tau] \mathcal{Z}_0(\chi_\tau)}{\mathcal{Z}_{\text{at}}(\lambda)} \frac{\delta[1 - g_0\lambda]}{\delta \lambda(\tau, 0)}. \end{aligned} \quad (93)$$

However, using the identity  $\det A = \exp[\text{Tr}(\ln A)]$  tells us that

$$\frac{\delta[1 - g_0\lambda]}{\delta \lambda(\tau, 0)} = -\det[1 - g_0\lambda] \{(1 - g_0\lambda)^{-1} g_0\}_{0,\tau}. \quad (94)$$

Substituting this result into the integral then gives

$$\chi_{\text{hyb}} = \int_0^\beta d\tau F(\tau) \int_0^\beta d\tau' [1 - g_0\lambda]_{0\tau'}^{-1} g_0(\tau', \tau), \quad (95)$$

where it should be noted that the auxiliary Green's function  $g_0$  is evaluated with the  $\chi_\tau$  field and therefore must be recalculated for each value of  $\tau$  in the integrand (since  $\chi_\tau$  varies with  $\tau$ ).

Now we introduce a Fourier transform for the  $\tau'$  variable to rearrange this result into the following final form:

$$\chi_{\text{hyb}} = \int_0^\beta d\tau F(\tau) T \sum_{mn} [1 - g_0\lambda]_{mn}^{-1} g_0(i\omega_n, \tau), \quad (96)$$

where the Green's function is Fourier transformed with respect to one coordinate only:

$$g_0(i\omega_n, \tau) = T \int_0^\beta d\tau' \exp(i\omega_n \tau') g_0(\tau', \tau). \quad (97)$$

The calculation of  $\chi_{\text{hyb}}$  requires little additional work to what is needed to calculate  $F(\tau)$ . At each value of  $\tau$  we need only invert the matrix  $[1 - g_0\lambda]$  and perform the relevant vector product with  $g_0$  and the matrix summation.

It is interesting to evaluate the susceptibility in the limit  $U \rightarrow 0$ . Here we have  $F(\tau) = -\exp[-\tau(E_f - \mu)] / \{1 + \exp[-\beta(E_f - \mu)]\}$ , and the auxiliary Green's function becomes time translation invariant; so the matrix is diagonal  $[1 - g_0\lambda]_{mn}^{-1} = (i\omega_m + \mu) / (i\omega_m + \mu - \lambda_m) \delta_{mn}$ , and the partial Fourier-transformed Green's function becomes  $g_0(i\omega_n, \tau) = \exp(i\omega_n \tau) / (i\omega_n + \mu)$ . The formula for the susceptibility in equation (96) can now be evaluated directly by performing the summation over the Matsubara frequencies to yield

$$\chi_{\text{hyb}}(U = 0) = \int d\epsilon \rho(\epsilon) \frac{f(\epsilon - \mu) - f(E_f - \mu)}{E_f - \epsilon}, \quad (98)$$

where  $f(x) = 1/[1 + \exp(\beta x)]$  is the Fermi function. As  $T \rightarrow 0$ , the susceptibility will diverge whenever the chemical potential is equal to  $E_f$  because the Fermi factors will

limit the integration to the region  $\epsilon \leq E_f$ , which will cause the integral to diverge logarithmically. A more careful analysis shows that the susceptibility will behave like

$$\chi_{\text{hyb}}(U = 0) \rightarrow -\frac{1}{2}\rho(E_f) \ln T + \text{constant}, \tag{99}$$

as  $T \rightarrow 0$ . Since we expect the susceptibility to be larger in the interacting case, this analysis is suggestive that the spontaneous hybridization will continue to diverge for non-zero  $U$  as well.

### § 5. NUMERICAL SOLUTIONS

The numerical implementation of the above procedure is depicted schematically in figure 2 and is described as follows. We start with an initial guess for the self-energy  $\Sigma^\sigma$  and calculate the local propagator in equation (2). Using equation (38) we calculate the bare atomic propagator  $G_{0n}^\sigma$  and find  $Z_0$  and  $Z_{\text{at}}$ . Next we obtain  $w_0, w_1$  and find  $G_n^\sigma$  from equation (37). Using  $G_{0n}^\sigma$  and  $G_n^\sigma$ , we compute the atomic self-energy and iterate the set of equations starting with the new self-energy until it converges to the fixed point.

The iterations on the imaginary axis give static properties, such as  $n_f$ , and the static spin and charge susceptibilities. Having found the f-electron filling  $w_1$  at each temperature, we iterate equations (2), (37) and (38) on the real axis and obtain the retarded dynamic properties, such as the spectral function, the resistivity, the magnetoresistance and the optical conductivity. At the fixed point, the spectral properties of the atom perturbed by the  $\lambda$  field coincide with the local spectral properties of the lattice.

#### 5.1. The f-electron spectrum and the results for classical intermediate-valence materials

As an example, we consider first the d-electron and the f-electron Green's functions for the spinless FK model on a hypercubic lattice, at half-filling. The interacting density of states for the conduction band is shown in figure 3, where  $\rho_d(\omega)$  is plotted versus frequency for several values of  $U$  in the high-temperature homogeneous phase. The interacting density of states is independent of temperature at high

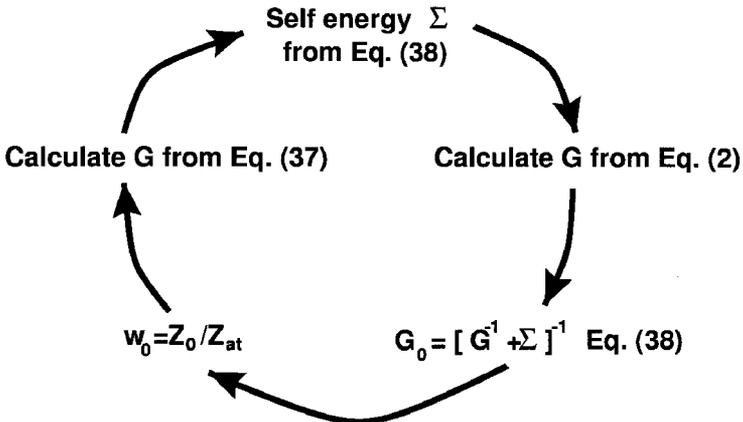


Figure 2. Iterative algorithm for determining the self-consistent solution of the local Green's function.

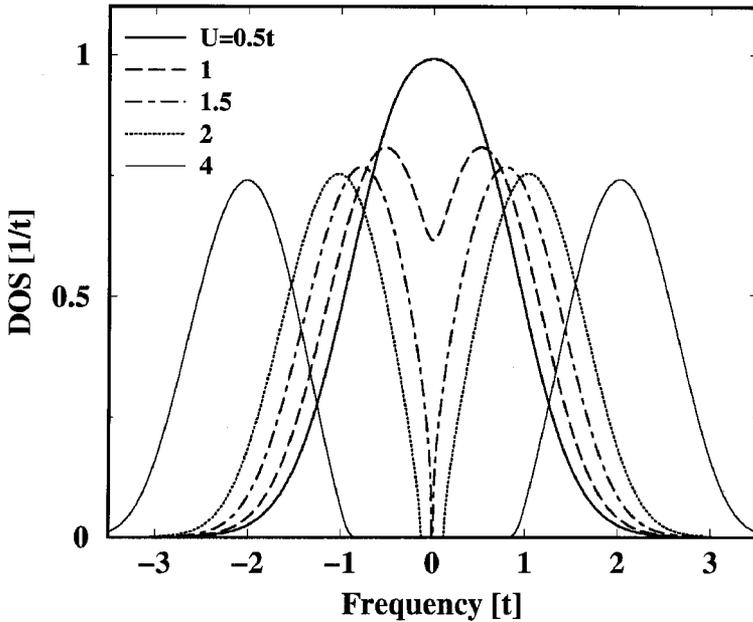


Figure 3. Interacting density of states DOS plotted versus  $\omega$  for several values of  $U$ , as indicated in the figure.

temperatures (van Dongen 1992) but not at low temperatures where the system undergoes a phase transition to an AB ordered ‘chessboard’ phase (Brandt and Mielsch 1989, Freericks and Lemanski 2000). A metal–insulator gap opens up at  $U = 1.5$ .

We have illustrated how to calculate the f-electron Green’s function, and the result for the spinless FK model on a hypercubic lattice at half-filling is plotted in figure 4 as a function of the Matsubara frequency, and for several values of  $U$ . In the limit  $U \rightarrow 0$ , the Green’s function becomes a non-interacting  $\delta$  function (which behaves like  $1/(i\omega_n + \mu - E_f)$  on the imaginary axis)

The sharp rise in the Green’s function can be clearly seen for small  $U$ . We expect that the Green’s function will become a correlated insulator at the same point that the conduction-electron Green’s function becomes insulating. Unfortunately, the f-electron Green’s function is temperature dependent here and, since we are working at finite temperature, we can only see the gap develop when  $U$  becomes large (this is easiest to see by the fact that  $F(i\omega_0) \rightarrow 0$ , which occurs for  $U$  larger than about 2.0).

Recent calculations on the spin- $\frac{1}{2}$  FK model on the Bethe lattice (Chung and Freericks 2000) have shown the existence of regions of parameter space where the ground state is not a charge-density wave state or a phase-separated state but remains homogeneous down to  $T = 0$ . In this region of the phase diagram, there are no other competing phases; so the system is eligible to have a  $T = 0$  divergence of the spontaneous hybridization. Here we illustrate this behaviour for the spinless model. Previous calculations have found the possibility of spontaneous hybridization to be precluded by other phases (Farkasovský, 1997, 1999, Czycholl 1999) or to occur for ‘singular’ density of states (Si *et al.* 1992). Here we provide numerical evidence for the divergence at  $T = 0$  in a restricted region of parameter space for ‘conventional lattices’.

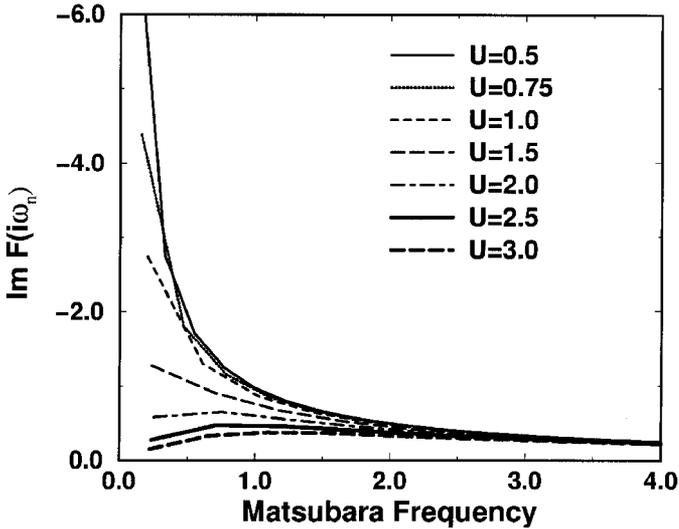


Figure 4. f-electron Green's function plotted versus Matsubara frequency, for several values of  $U$  as indicated in the figure.

We begin by finding a region of parameter space in the spinless FK model where the classical intermediate-valence state is stable against phase separation all the way down to  $T = 0$ . This is simplest to find by repeating the previous analysis for the spin- $\frac{1}{2}$  case (Chung and Freericks 2000) (we perform calculations on the Bethe lattice here). We can show that, if we choose  $n_{\text{tot}} = 0.5$  and  $-1 < E_f < 0$ , then the intermediate-valence state is stable for small  $U$ . We choose  $E_f = -0.75$ , where the intermediate-valence state appears to be stable for all values of  $U$  (we did not perform a Maxwell construction to check for first-order phase transitions). A plot of the average f-electron concentration and the uniform charge susceptibility versus temperature is given in figure 5.

Note how the f-electron concentration remains finite for all  $U$  as  $T \rightarrow 0$  and how the compressibility remains finite as well. The inverse of the hybridization susceptibility is shown in figure 6. Note how the logarithmic divergence at  $U = 0$  is difficult to see in this figure and how the inverse susceptibility decreases as  $U$  increases. This then suggests that the susceptibility will continue to diverge at  $T = 0$  for all  $U$  (we know from Elitzur's theorem that it cannot diverge at any finite  $T$ ).

### 5.2. Results for $\text{YbInCu}_4$

The numerical results for the spin- $\frac{1}{2}$  FK model exhibit several features that one finds in the family of  $\text{YbInCu}_4$  compounds. We consider here a hypercubic lattice with a total electron filling of 1.5 and several values of  $E_f$  and  $U$ , such that  $E_f > \mu(T = 0)$ , since that is the regime where the f occupation can change rapidly as a function of  $T$ .

The main results can be summarized in the following way. The occupancy of the f holes at high temperatures is large and there is a huge magnetic degeneracy. The f holes are energetically unfavourable but are maintained because of their large magnetic entropy. In figure 7 we show  $n_f$  as a function of temperature, plotted for  $U = 4$ , and  $E_f$  ranging from  $-0.2$  to  $-0.7$ . Below a certain temperature, which depends on  $U$

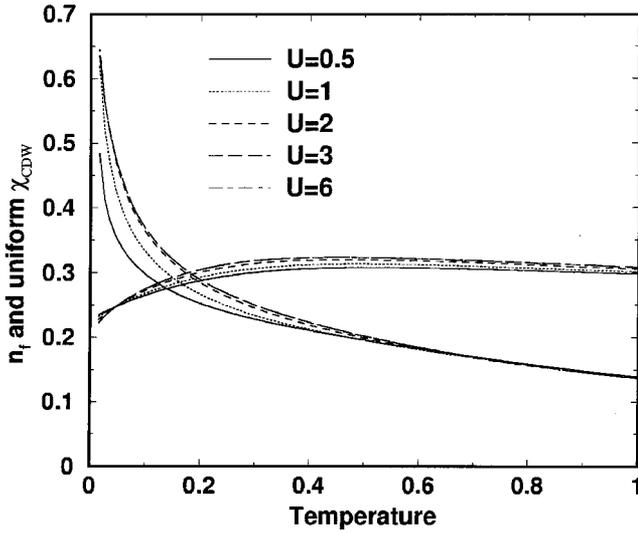


Figure 5. The average f-electron concentration and the uniform charge susceptibility plotted versus temperature, for several values of  $U$  as indicated in the figure. Both quantities vary little with the interaction strength. The f-electron concentration is the set of curves that decrease as  $T \rightarrow 0$  and the susceptibility is the set of curves that increase as  $T \rightarrow 0$ .

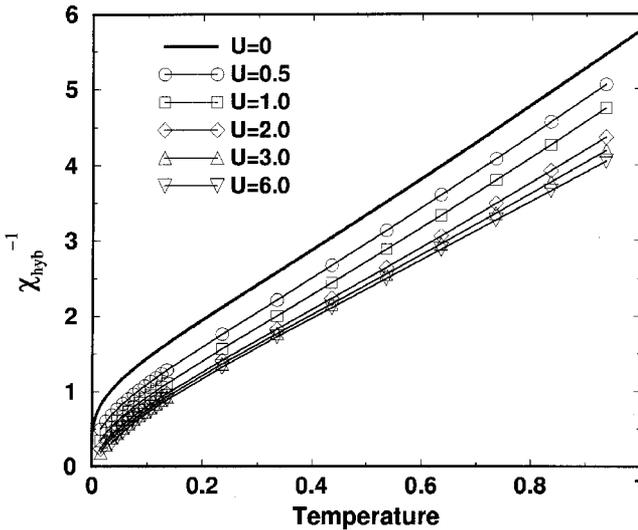


Figure 6. The inverse of the hybridization susceptibility plotted versus temperature, for several values of  $U$  as indicated in the figure.

and  $E_f$ , there is a rapid transition from the high-temperature phase with a moderate f occupancy to the low-temperature phase where  $n_f \rightarrow 0$ . The ‘transition’ occurs at a crossover temperature  $T_V$  and becomes sharper and is pushed to lower temperatures as  $E_f$  decreases at fixed  $U$ . The uniform f-spin susceptibility is obtained by calculating the spin–spin correlation function (Freericks and Zlatić, 1998, Zlatić and

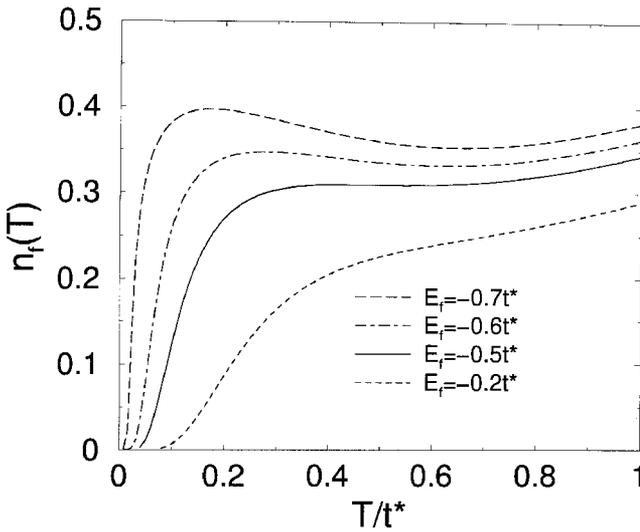


Figure 7. Number of f holes plotted versus  $T$  for  $U = 4$ .  $E_f$  increases from top to bottom and equals  $-0.7$ ,  $-0.6$ ,  $-0.5$ , and  $-0.2$ , respectively.

Freericks 2001) and is given by  $\chi(T) = Cn_f(T)/T$ , where  $C = g_L^2 \mu_B^2 J(J+1)/3k_B$  is the Curie constant. Thus it is clear that, in the high-temperature phase, where  $n_f(T)$  does not change much, the susceptibility approaches the Curie law. However as the f-d interaction gives rise to changes in the f occupancy, the susceptibility assumes only the form of an approximate Curie-Weiss law. In the region where  $n_f(T)$  changes rapidly, the susceptibility exhibits a sharp maximum, which separates the magnetic and non-magnetic regions of the phase diagram. Below  $T_v$ , the f susceptibility is negligibly small, and the total susceptibility is due to the conduction electrons and is Pauli like. The other static correlation functions have also been calculated, and the results obtained in the homogeneous phase have been discussed by Freericks and Zlatić (1998) and Zlatić and Freericks (2001).

The interacting density of states,  $\rho_d(\omega)$  for the conduction electrons, calculated for  $U = 4$  and  $E_f = -0.5$ , is plotted in figure 8 versus frequency for several temperatures. (The zero of energy is measured with respect to  $\mu$ .) The high-temperature density of states has a gap of the order of  $U$ , and the chemical potential is located within the gap. Below the crossover temperature  $T_v$ ,  $n_f$  is small, the correlation effects are reduced, and  $\rho_d(\omega)$  assumes a nearly non-interacting shape, with large  $\rho_d(\mu)$  and half-width  $W \approx 1$ .

The intraband optical conductivity is determined by an integral of the spectral function (Pruschke *et al.* 1995) as

$$\sigma(\omega) = \sigma_0 \int d\epsilon \rho(\epsilon) \int d\nu \frac{f(\nu) - f(\nu + \omega)}{\omega} A(\epsilon, \nu) A(\epsilon, \nu + \omega), \quad (100)$$

where  $A(\epsilon, \nu) = -(1/\pi) \text{Im} [1/(\nu + \mu - \Sigma(\nu) - \epsilon)]$  is the spectral function. The result for  $\sigma(\omega)$  obtained in such a way is plotted in figure 9 as a function of frequency, for several temperatures. Above  $T_v$ , we observe a reduced Drude peak at around  $\omega = 0$  and a pronounced high-frequency peak around  $\omega \approx U$ . The shape of  $\sigma(\omega)$  changes completely across  $T_v$ . Below  $T_v$  the Drude peak is fully developed and there is no

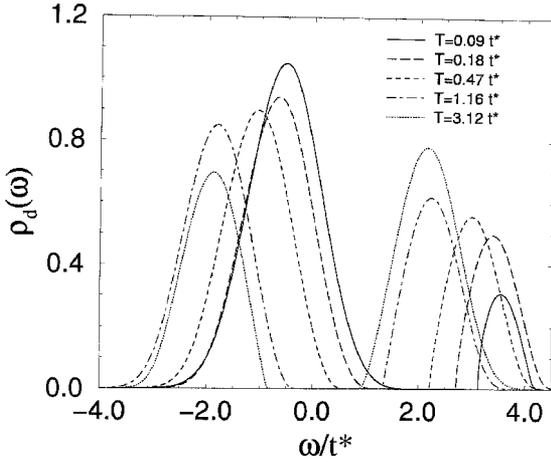


Figure 8. Interacting density of states plotted versus  $\omega$  for  $U = 4$  and  $E_f = -0.5$  ( $T_v = 0.14$ ), and for various temperatures, as indicated in the figure.

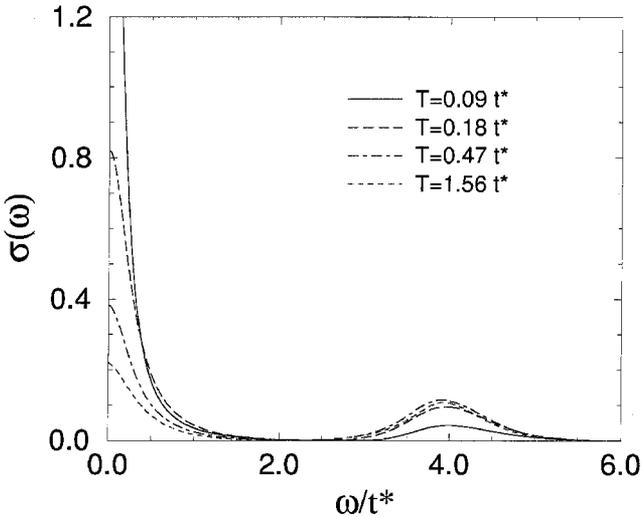


Figure 9. Optical conductivity plotted versus  $\omega/t^*$  for various temperatures.  $U$ ,  $E_f$  and  $T_v$ , are the same as in figure 8.

high-energy (intraband) structure. However, if the renormalized f level is close to  $\mu$ , the interband d–f transition could lead to an additional mid-infrared peak.

If we estimate the f–d correlation in  $\text{YbInCu}_4$  from the  $8000\text{ cm}^{-1}$  peak in the optical conductivity data (Garner *et al.* 2000), we obtain the experimental value  $U \approx 1\text{ eV}$ . Together with  $T_v = 42\text{ K}$  (Sarrao 1999) this gives the ratio  $U/T_v \approx 200$ . If we take  $U = 4$  and adjust  $E_f$  so as to bring the theoretical value of  $T_v$  into agreement with the thermodynamic and transport data on  $\text{YbInCu}_4$ , we obtain a high-frequency peak in  $\sigma(\omega)$  at about  $8000$ ,  $6000$  and  $1500\text{ cm}^{-1}$ , for  $E_f = -0.75$ ,  $-0.7$  and  $-0.5$ , respectively.

From the preceding discussion it is clear that the FK model captures the main features of the experimental data for YbInCu<sub>4</sub> and similar compounds. However, our calculations describe much better the doped Yb systems with broad transitions than those compounds which show a first-order transition. The numerical curves can be made sharper (by adjusting the parameters) but they only become discontinuous in a narrow parameter range. Our results indicate that the temperature- and field-induced anomalies are related to a metal–insulator transition, which is caused by a large FK interaction and triggered by the temperature- or the field-induced change in the f occupancy.

## § 6. SUMMARY AND OUTLOOK

We studied the static and dynamic correlation functions of the infinite-dimensional FK model by an EOM method. The exact solution (Brandt and Mielsch 1989, Brandt and Urbanek 1992) has been presented for the model with an arbitrary number of electrons, and for a  $(2s + 1)$ -fold degenerate d state and a  $(2S + 1)$ -fold degenerate f state. In the large- $U$  limit, and for a range of parameters, the spin- $\frac{1}{2}$  model exhibits a transition from a high-temperature semiconductor or a semimetal, with well-defined f moments, to a low-temperature Pauli metal. The static and dynamic correlation functions show many similarities with the experimental data on valence fluctuating Yb compounds, and perhaps on SmB<sub>6</sub>, but the crossovers calculated for the spin- $\frac{1}{2}$  model are less sharp than seen in the experimental data. We believe that the sharpness of the transition in Yb compounds is due to the large entropy change, when Yb ions switch from the magnetic  $4f^{13}$  configuration (with an eight-fold degenerate f hole in the  $J = \frac{7}{2}$  spin–orbit state) to a non-magnetic  $4f^{14}$  configuration. The model with an eight-fold degenerate f state and a two-fold degenerate d state can easily be solved by the methods explained in this paper, and we expect to see a much sharper transition there. The numerical analysis of such a model and the study of the correlation functions for various values of the ratio  $(2S + 1)/(2s + 1)$  will be the subject of subsequent work.

A more serious difficulty with the FK model is that it neglects the quantum fluctuations of the f state and considers only statistical fluctuations. That is, the lifetime of an f state is assumed to be infinite, and the width of the f spectrum arises only because the f electrons couple to density fluctuations in the conduction band. Thus, the valence transition in the FK model is accompanied by a substantial change in the f occupancy, and the loss of the magnetic moment is associated with the loss of f holes. In real materials, the loss of the local moment seems to be due to quantum fluctuations and lifetime effects, rather than to the disappearance of the f holes. The description of the quantum valence-fluctuating ground state would have to consider the hybridization between the f and d states, and that would require a periodic Anderson model with an additional FK interaction. The EOM method elaborated in this paper does not produce the solution for such a generalized model.

The actual situation pertaining to Yb ions in the mixed-valence state might be quite complicated, since one must consider an extremely asymmetric limit of the Anderson model, in which the ground state is not Kondo like, there is no Kondo resonance and there is no single universal energy scale which is relevant at all temperatures (Krishnamurti *et al.* 1980). We speculate that the periodic Anderson model with a large FK term will exhibit the same behaviour as the FK model at high temperatures. Indeed, if the conduction band and the f level are gapped, and the width of the f level is large, then the effect of the hybridization can be accounted for

by renormalizing the parameters of the FK model. On the other hand, if the low-temperature state of the full model is close to the valence-fluctuating fixed point with the conduction band and hybridized f level close to the Fermi level, then the likely effect of the FK correlation is to renormalize the parameters of the Anderson model.

This picture is borne out from our examination of the spontaneous hybridization for the spinless FK model on the Bethe lattice. We find that, as  $T$  is lowered, the system seems to have a logarithmic divergence in the spontaneous hybridization susceptibility at  $T = 0$ . Normally we cannot reach such a phase because the system will have a phase transition to either a phase-separated state or a charge-density wave, but we can tune the system so that it remains in a classical intermediate-valence state down to  $T = 0$ . When this occurs, the effects of even a small hybridization will take the system away from the FK fixed point at low enough  $T$  and hybridization can no longer be neglected.

#### ACKNOWLEDGEMENTS

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#### APPENDIX A

The matrix elements in equations (87) and (88), which are used to calculate the determinant in equation (89) for the f propagator, are given by the expressions

$$M_{(n \neq m)} = \xi_0 A_{nm} + (\xi_0 - 1) B_{nm}, \tag{A 1}$$

$$M_{(n,n)} = \xi_0 C_{nn} + (\xi_0 - 1) D_{nn}, \tag{A 2}$$

where  $\xi_0 = 1/[1 + \exp(U\tau - \beta\mu)]$ , and

$$\begin{aligned} A_{mn} = & \frac{1}{i(\omega_m - \omega_n)(i\omega_m + \mu - U)} - \frac{\exp(i\tau\omega_n + \mu\tau - \beta\mu)}{(i\omega_m + \mu)(i\omega_n + \mu)} \\ & + \frac{\{\exp[-(i\omega_m + \mu)\tau] + \exp(-\beta\mu)\}\{\exp[(i\omega_n + \mu)\tau] - \exp(U\tau)\}}{(i\omega_m + \mu)(i\omega_n + \mu - U)} \\ & - \frac{1}{i(\omega_m - \omega_n)(i\omega_n + \mu)} + \frac{\exp[-i(\omega_m - \omega_n)\tau]}{i(\omega_m - \omega_n)(i\omega_m + \mu)} \\ & + \frac{\exp[-(i\omega_m + \mu)\tau]\{i(\omega_m - \omega_n)\exp(U\tau) - (i\omega_m + \mu - U)\exp[(i\omega_n + \mu)\tau]\}}{i(\omega_m - \omega_n)(i\omega_m + \mu - U)(i\omega_n + \mu - U)}, \end{aligned} \tag{A 3}$$

$$\begin{aligned}
B_{mn} = & \frac{-1}{i(\omega_m - \omega_n)(i\omega_n + \mu - U)} - \frac{\exp[-i(\omega_m - \omega_n)\tau]}{i(\omega_m - \omega_n)(i\omega_n + \mu)} \\
& - \frac{\exp(i\omega_n\tau) \{ \exp(-i\omega_m\tau) - \exp[(\mu - U)\tau] \}}{(i\omega_m + \mu - U)(i\omega_n + \mu)} \\
& - \frac{\exp(\beta\mu) \{ \exp[(-i\omega_m + \mu)\tau] - \exp(-U\tau) \}}{(i\omega_m + \mu - U)(i\omega_n + \mu)} \\
& + \frac{i\omega_n + \mu - i(\omega_m - \omega_n) \exp[\beta\mu - (i\omega_m + \mu)\tau]}{i(\omega_m - \omega_n)(i\omega_m + \mu)(i\omega_n + \mu)} \\
& + \frac{\exp[-i(\omega_m - \omega_n)\tau] \{ i\omega_n + \mu - U + i(\omega_m - \omega_n) \exp[(i\omega_m + \mu - U)\tau] \}}{i(\omega_m - \omega_n)(i\omega_m + \mu - U)(i\omega_n + \mu - U)}, \quad (\text{A } 4)
\end{aligned}$$

$$\begin{aligned}
C_{mm} = & \frac{(i\omega_n + \mu - U)\tau - 1 + \exp[-(i\omega_n + \mu - U)\tau]}{(i\omega_n + \mu - U)^2} \\
& + \frac{1 - \exp[-(i\omega_n + \mu - U)\tau] - \exp(-\beta\mu) \{ \exp[(i\omega_n + \mu)\tau] - \exp(U\tau) \}}{(i\omega_n + \mu)(i\omega_n + \mu - U)} \\
& + \frac{(i\omega_n + \mu)(\beta - \tau) - 1 - \exp[-\beta\mu + (i\omega_n + \mu)\tau]}{(i\omega_n + \mu)^2}, \quad (\text{A } 5)
\end{aligned}$$

$$\begin{aligned}
D_{mm} = & \frac{-(i\omega_n + \mu - U)\tau - 1 + \exp[(i\omega_n + \mu - U)\tau]}{(i\omega_n + \mu - U)^2} \\
& + \frac{1 - \exp[(i\omega_n + \mu - U)\tau] - \exp(\beta\mu) \{ \exp[-(i\omega_n + \mu)\tau] - \exp(-U\tau) \}}{(i\omega_n + \mu)(i\omega_n + \mu - U)} \\
& + \frac{(i\omega_n + \mu)(\tau - \beta) - 1 - \exp[\beta\mu - (i\omega_n + \mu)\tau]}{(i\omega_n + \mu)^2}. \quad (\text{A } 6)
\end{aligned}$$

These expressions generalize the matrix elements obtained by Brandt and Urbanek (1992) for the system with electron–hole symmetry. We checked that for  $\mu = U/2$  our expressions agree with those of Brandt and Urbanek (1992) for  $m \neq n$ , but we find a slight inconsistency for the case  $m = n$ . The formulae given here, and those given by Brandt and Urbanek agree only if the minus sign in front of the  $i\omega_m$  term which appears in the numerator of the first line of the expression for  $M_{(m,m)}$  in the paper by Brandt and Urbanek (1992) is replaced by a plus sign.

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