

Continuous-time impurity solvers

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I. QUANTUM IMPURITY MODELS

An quantum impurity model describes an atom or molecule embedded in some host or *bath*, with which it can exchange electrons. This exchange of electrons allows the impurity to make transitions between different quantum states, and leads to a non-trivial dynamics. Quantum impurity models play a prominent role in nano-science as representations of quantum dots or molecular conductors. They also serve as auxiliary problems, whose solution yields the “dynamical mean field” description of correlated lattice models.

The Hamiltonian of a quantum impurity model contains three terms,

$$H = H_{\text{loc}} + H_{\text{bath}} + H_{\text{mix}}. \quad (1)$$

H_{loc} describes the impurity, characterized by a small number of degrees of freedom (typically spin and orbital degrees of freedom, which we denote by a, b, \dots), H_{bath} describes an infinite reservoir of free electrons, labeled by a continuum of quantum numbers p and a discrete set of quantum numbers ν (e. g. spin). H_{mix} describes the exchange of electrons between the impurity and the bath in terms of hybridization amplitudes $V_p^{a\nu}$. Explicitly, the three terms are

$$H_{\text{loc}} = \sum_{ab} E^{ab} d_a^\dagger d_b + \sum_{abcd} U^{abcd} d_a^\dagger d_b^\dagger d_c d_d, \quad (2)$$

$$H_{\text{bath}} = \sum_{p,\nu} \epsilon_p^b a_{p,\nu}^\dagger a_{p,\nu}, \quad (3)$$

$$H_{\text{mix}} = \sum_{p,a,\nu} (V_p^{a,\nu} d_a^\dagger a_{p,\nu} + h.c.). \quad (4)$$

We will concentrate in the following on the “one-orbital Anderson model” (AIM), a simplified representation of a magnetic impurity in a metallic host. In the AIM, the discrete quantum number labelling the impurity or bath state is the spin σ , so the Hilbert space of

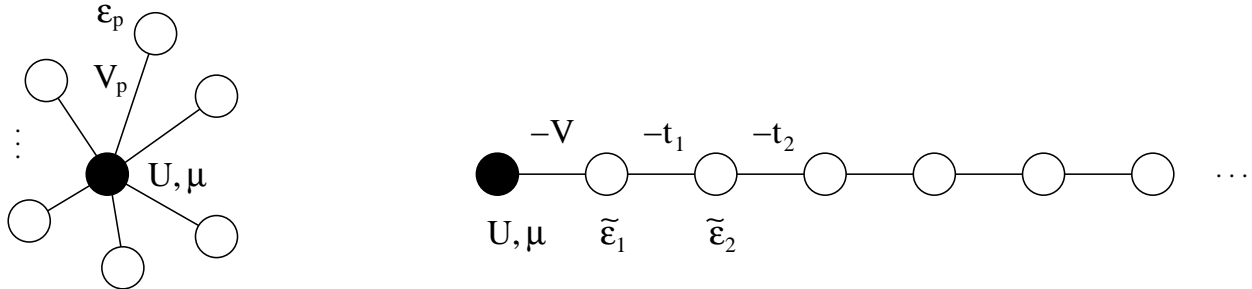


FIG. 1: Left: Schematic representation of an AIM. Spin up and down electrons on the impurity (black dot) interact with an on-site energy U and can hop to a continuum of non-interacting bath levels with energy ϵ_p . The amplitudes for these transitions are given by the hybridization parameters V_p^* . Right: Chain representation of the AIM. All the hopping parameters can be chosen negative by a suitable gauge transformation.

the local problem $H_{\text{loc}} = H_0 + H_U$,

$$H_0 = -\mu(n_\uparrow + n_\downarrow), \quad (5)$$

$$H_U = Un_\uparrow n_\downarrow, \quad (6)$$

has dimension four, and the bath and mixing terms are given by

$$H_{\text{bath}} = \sum_{p,\sigma} \epsilon_p a_{p,\sigma}^\dagger a_{p,\sigma}, \quad (7)$$

$$H_{\text{mix}} = \sum_{p,\sigma} (V_p^\sigma d_\sigma^\dagger a_{p,\sigma} + h.c.). \quad (8)$$

An illustration of the AIM is shown in the left hand panel of Fig. 1.

A. Chain representation

The AIM (5)-(8) can be mapped onto a semi-infinite chain, with the first site corresponding to the impurity. This mapping corresponds to a transformation of the operators $(d, a_{p_1}, a_{p_2}, \dots)$ to new operators (d, c_1, c_2, \dots) such that $H_0 + H_{\text{bath}} + H_{\text{mix}}$ becomes tridiagonal.

$$\begin{pmatrix} -\mu & V_{p_1} & V_{p_2} & V_{p_3} & \dots \\ V_{p_1}^* & \epsilon_{p_1} & & & \\ V_{p_2}^* & & \epsilon_{p_2} & & \\ V_{p_3}^* & & & \epsilon_{p_3} & \\ \dots & & & & \dots \end{pmatrix} \rightarrow \begin{pmatrix} -\mu & -V & & & \\ -V & \tilde{\epsilon}_1 & -t_1 & & \\ & -t_1 & \tilde{\epsilon}_2 & -t_2 & \\ & & -t_2 & \tilde{\epsilon}_3 & \dots \\ & & & & \dots \dots \end{pmatrix} \quad (9)$$

In the chain representation, the impurity orbital remains unchanged, and the hopping term from the impurity to the first site of the chain is given by $V = (\sum_p |V_p|^2)^{1/2}$. The phase factors in this transformation can be chosen such that all the hopping terms are positive ($V > 0$, $t_i > 0$, $i = 1, 2, \dots$).

B. Action formulation

We may integrate out the bath degrees of freedom and express the partition function of the AIM as

$$Z = \text{Tr} T e^{-S} \quad (10)$$

with action $S = S_{\text{hyb}} + S_{\text{loc}}$ given by

$$S_{\text{hyb}} = \sum_{\sigma} \int_0^{\beta} d\tau d\tau' d_{\sigma}^{\dagger}(\tau) \Delta_{\sigma}(\tau - \tau') d_{\sigma}(\tau'), \quad (11)$$

$$S_{\text{loc}} = \int_0^{\beta} d\tau [-\mu(n_{\uparrow}(\tau) + n_{\downarrow}(\tau)) + U n_{\uparrow}(\tau) n_{\downarrow}(\tau)]. \quad (12)$$

The hybridization function $\Delta_{\sigma}(\tau - \tau')$ in Eq. (11) represents the amplitude for hopping from the impurity into the bath at time τ' and back onto the impurity at time τ . It is a function of the bath level energies and hybridization amplitudes, which is most conveniently expressed in frequency space ($i\omega_n = (2n + 1)\pi/\beta$):

$$\Delta_{\sigma}(i\omega_n) = \sum_p \frac{|V_p^{\sigma}|^2}{i\omega_n - \epsilon_p}. \quad (13)$$

It is also sometimes useful to introduce the noninteracting bath Green's function G_0 defined as

$$G_{0,\sigma}^{-1}(i\omega_n) = i\omega_n + \mu - \Delta_{\sigma}(i\omega_n). \quad (14)$$

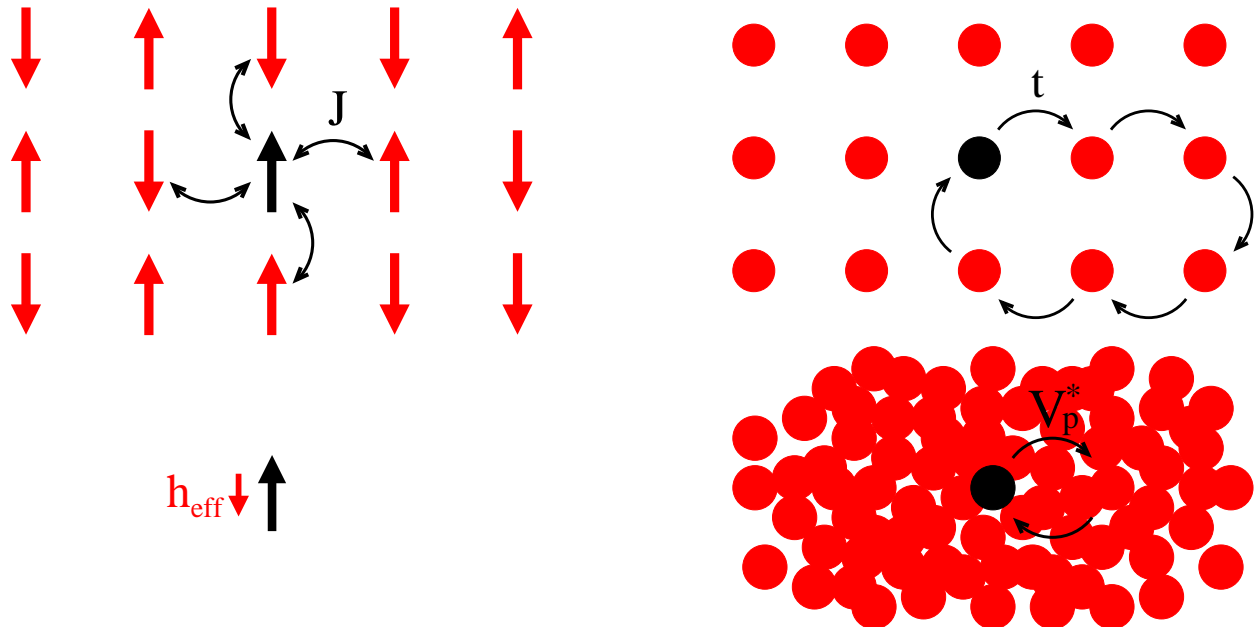


FIG. 2: Mapping of the classical Ising model to a single-site effective model in static mean field theory (left panel) and mapping of the Hubbard model to a quantum impurity model in dynamical mean field theory (right panel).

II. DYNAMICAL MEAN FIELD THEORY

Quantum impurity models are a key ingredient in the dynamical mean field (DMFT) formalism, which provides an approximate description of correlated lattice models. The success of DMFT has created a demand for powerful and flexible impurity solvers and triggered the development of the continuous-time solvers discussed in this chapter. We will thus briefly introduce the DMFT approximation, which maps an interacting lattice model, such as the one-band Hubbard model onto an effective *single-site* problem plus a self-consistency condition.

To appreciate the basic strategy, it is useful to briefly recall the static mean-field approximation of the classical Ising model, which is illustrated in the left hand panel of Fig. 2. Here, one focuses on a particular spin of the lattice, S_0 , and replaces the remaining degrees of freedom by an effective external magnetic field $h_{\text{eff}} = zJm$ (z is the coordination number and m the magnetization per site). The lattice system

$$H^{\text{Ising}} = -J \sum_{\langle ij \rangle} S_i S_j \quad (15)$$

is thus mapped to the single site effective model

$$H_{\text{eff}}^{\text{Ising}} = -h_{\text{eff}} S_0. \quad (16)$$

From this single-site model we can easily compute the magnetization

$$m_{\text{eff}} = \tanh(\beta h_{\text{eff}}), \quad (17)$$

and identifying the magnetization m of the lattice problem with the magnetization m_{eff} of the single site effective model yields the self-consistency condition

$$m \equiv m_{\text{eff}} = \tanh(\beta z J m). \quad (18)$$

We now turn to the Hubbard model,

$$H_{\text{Hubbard}} = -\mu \sum_{i,\sigma} n_{i,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + t \sum_{\langle i,j \rangle \sigma} a_{i,\sigma}^\dagger a_{j,\sigma} \quad (19)$$

which describes electrons hopping between nearest neighbor sites of some lattice with amplitude t and interacting on-site with energy U . Pursuing a similar strategy as in the Ising case, we focus on one particular site (black in the right hand panels of Fig. 2) of the lattice and replace the remaining degrees of freedom of the model by a bath of non-interacting levels. The single-site effective problem thus becomes an AIM,

$$H_{\text{imp}} = -\mu(n_\uparrow + n_\downarrow) + U n_\uparrow n_\downarrow + \sum_{p,\sigma} (V_p^\sigma d_\sigma^\dagger a_{p,\sigma} + h.c.) + \sum_{p,\sigma} \epsilon_p a_{p,\sigma}^\dagger a_{p,\sigma}. \quad (20)$$

Here, the d^\dagger create electrons on the ‘‘impurity’’, $n = d^\dagger d$ and the a_p^\dagger create bath states labeled by a quantum number p . The energy levels of the bath are ϵ_p . Processes where an electron hops from the black site into the lattice and after some excursion through the lattice returns to its original place (top right panel of Fig. 2) are represented in the single-site effective model by transitions from the impurity into the bath and back (bottom right panel of Fig. 2). The amplitudes for such transitions are given by the hybridization parameters V_p of the impurity model and the task will be to optimize the parameters ϵ_p and V_p in such a way that the bath mimics the lattice environment.

More specifically, if $G(k, i\omega_n)$ is the momentum dependent lattice Green function, we would like to determine bath parameters such that the impurity Green function $G_{\text{imp}}(i\omega_n)$ reproduces the *local* (momentum averaged) lattice Green function,

$$\sum_k G(k, i\omega_n) \equiv G_{\text{imp}}(i\omega_n). \quad (21)$$

A. DMFT approximation

The solution to Eq. (21) is computed iteratively and these DMFT iterations involve as the essential approximation of the method a simplification of the momentum-dependence of the lattice self-energy. The self-energy describes the effect of interactions on the propagation of electrons. In the non-interacting model ($U = 0$) the lattice Green function is given by $G_{U=0}(k, \omega) = [i\omega_n + \mu - \epsilon_k]^{-1}$, with ϵ_k the Fourier transform of the hopping matrix, whereas the Green function of the interacting model is given by $G(k, \omega) = [i\omega_n + \mu - \epsilon_k - \Sigma(k, i\omega_n)]^{-1}$. Therefore $\Sigma(k, i\omega_n) = G_{U=0}^{-1}(k, i\omega_n) - G(k, i\omega_n)^{-1}$, and similarly the impurity self-energy is given by $\Sigma_{\text{imp}}(i\omega_n) = G_{\text{imp}, U=0}^{-1}(i\omega_n) - G_{\text{imp}}(i\omega_n)^{-1}$. The DMFT approximation amounts to identifying the lattice self-energy with the momentum-independent impurity self-energy,

$$\Sigma(k, i\omega_n) \approx \Sigma_{\text{imp}}(i\omega_n). \quad (22)$$

Using this approximation we can rewrite Eq. (21) as

$$\sum_k [i\omega_n + \mu - \epsilon_k - \Sigma_{\text{imp}}(i\omega_n)]^{-1} \equiv G_{\text{imp}}(i\omega_n). \quad (23)$$

Since both $G_{\text{imp}}(i\omega_n)$ and $\Sigma_{\text{imp}}(i\omega_n)$ depend on the impurity model parameters ϵ_p and V_p we obtain a self-consistency condition for the impurity model.

B. DMFT self-consistency loop

Starting from an arbitrary initial bath Green function $G_0(i\omega_n)$ (for example the solution of the noninteracting problem), one iterates the following steps until convergence is reached:

1. Solve the impurity problem, i. e. compute the Green function $G_{\text{imp}}(i\omega_n)$ for the given “bath” $G_0(i\omega_n)$,
2. Extract the self-energy of the impurity model: $\Sigma_{\text{imp}}(i\omega_n) = G_0^{-1}(i\omega_n) - G_{\text{imp}}^{-1}(i\omega_n)$,
3. Identify the lattice self-energy with the impurity self-energy ($\Sigma(k, i\omega_n) = \Sigma_{\text{imp}}(i\omega_n)$) and compute the local lattice Green function $G_{\text{loc}}(i\omega_n) = \sum_k [i\omega_n + \mu - \Sigma_{\text{imp}}(i\omega_n)]^{-1}$,
4. Apply the DMFT self-consistency condition ($G_{\text{loc}}(i\omega_n) = G_{\text{imp}}(i\omega_n)$) and use this to define a new “bath” $G_{0,\text{new}}^{-1}(i\omega_n) = G_{\text{loc}}^{-1}(i\omega_n) + \Sigma_{\text{imp}}(i\omega_n)$.

The computationally expensive step in this procedure is the solution of the impurity problem. Once the calculation has converged, the bath contains information about the topology of the lattice. The impurity, which exchanges electrons with the bath, will thus feel (at least to some extent) as if it were a site of the lattice. Obviously, not all the physics can be captured by a single-site impurity model. In particular spatial fluctuations, which become important in low-dimensional systems, are completely neglected. Dynamical mean field calculations become exact in the limit of infinite dimension or coordination number, in the non-interacting limit ($U = 0$) and in the atomic limit ($t = 0$).

C. Simulation of strongly correlated materials

The DMFT formalism can describe band-like behavior (quasi-particle peaks) *and* atomic-like behavior (Hubbard bands). It thus captures the competition between localization and delocalization which plays a crucial role in the physics of strongly correlated materials. In order to enable “ab-initio” simulations of real compounds, the dynamical mean field framework can be combined with density functional theory in the LDA approximation. The resulting formalism is called “LDA+DMFT”. The idea is to use the Kohn-Sham eigenvalues ϵ_k^{KS} in the self-consistency equation (23) instead of the dispersion of some tight-binding model. However, by doing so one encounters a problem: while density functional theory in the LDA approximation does not take into account all the correlation effects it captures some of them. If we now explicitly describe the local interaction in the strongly correlated orbitals via some U -term in an effective impurity model, some interaction contributions appear twice and this “double counting” of interaction energy must be compensated by adding a *double counting correction* E_{DC} to the self-energy of the correlated orbitals. The self-consistency condition thus becomes

$$\sum_k [i\omega_n + \mu - \epsilon_k^{KS} - \Sigma_{\text{imp}}(i\omega_n) - E_{DC}]^{-1} \equiv G_{\text{imp}}(i\omega_n). \quad (24)$$

There is no clean and consistent solution to the double counting problem in LDA+DMFT. In practice, one uses formulas like $E_{DC} = U\langle n \rangle$ with $\langle n \rangle$ the average occupancy.

Most material simulations will involve several bands, so that the Kohn-Sham eigenvalues define a matrix H_k^{LDA} in Wannier orbital space. Only the d - or f -orbitals will be explicitly treated in the impurity calculation and yield a self-energy. Thus Σ_{imp} will be a matrix of

the same size as H_k^{LDA} , but the only non-zero elements will be in the block corresponding to the strongly correlated d - or f -orbitals. Similarly the double-counting correction will be a diagonal matrix with non-zero elements only for the correlated orbitals. In the multi-orbital case, one can use as double counting correction $E_{DC} = \langle U \rangle \langle n_{\text{corr}} \rangle$, with $\langle U \rangle$ the average of the interaction parameters of the (multi-orbital) impurity problem and $\langle n_{\text{corr}} \rangle$ the average occupancy of the correlated orbitals. This orbital-independent shift assures that the crystal-field splittings in the LDA band structure are preserved by the double counting correction. The chemical potential μ is adjusted such that the correct total number of electrons in the correlated and uncorrelated orbitals is obtained.

III. CONTINUOUS-TIME IMPURITY SOLVERS - GENERAL RECIPE

Quantum impurity models are 0+1 dimensional quantum field theories and as such computationally much more tractable than interacting lattice models. By “solving the impurity model” we essentially mean computing the impurity Green function ($0 < \tau < \beta$)

$$G(\tau) = \langle T d(\tau) d^\dagger(0) \rangle = \frac{1}{Z} \text{Tr} \left[e^{-(\beta-\tau)H} d e^{-\tau H} d^\dagger \right], \quad (25)$$

with $Z = \text{Tr}[e^{-\beta H}]$, the impurity model partition function, β the inverse temperature, and $\text{Tr} = \text{Tr}_d \text{Tr}_a$ the trace over the impurity and bath states.

Hamiltonian based methods such as exact diagonalization or numerical RG explicitly treat a finite number of bath states, and are suitable for single orbital models. However, because the number of bath states must be increased proportional to the number of orbitals, the computational effort grows exponentially with system size, and requires severe truncations of the bath already for two orbitals. Monte Carlo methods have the advantage that the bath is integrated out and thus the (infinite) size of the bath Hilbert space does not affect the simulation. While restricted to finite temperature, Monte Carlo methods are thus the method of choice for the solution of large multi-orbital or cluster impurity problems.

We will discuss here two complementary continuous-time Monte Carlo techniques: (i) a weak-coupling approach, which scales favorably with system size and allows the efficient simulation of large impurity clusters, and (ii) a strong-coupling approach, which can handle impurity models with strong interactions. For simplicity, we will focus on the single-orbital AIM impurity model defined in Eqs. (5)-(8).

Continuous-time Monte Carlo algorithms are based on an expansion of the partition function into a series of “diagrams” and the stochastic sampling of (collections) of these diagrams. We represent the partition function as a sum (or, more precisely, integral) of configurations c with weight w_c ,

$$Z = \sum_c w_c, \quad (26)$$

and implement a random walk $c_1 \rightarrow c_2 \rightarrow c_3 \rightarrow \dots$ in configuration space in such a way that each configuration can be reached from any other in a finite number of steps (*ergodicity*) and that *detailed balance* is satisfied,

$$|w_{c_1}|p(c_1 \rightarrow c_2) = |w_{c_2}|p(c_2 \rightarrow c_1). \quad (27)$$

This assures that each configuration is visited with a probability proportional to $|w_c|$ and one can thus obtain an estimate for the Green function from a finite number N of measurements:

$$G = \frac{\sum_c w_c G_c}{\sum_c w_c} = \frac{\sum_c |w_c| \text{sign}_c G_c}{\sum_c |w_c| \text{sign}_c} = \frac{\langle \text{sign} \cdot G \rangle_{MC}}{\langle \text{sign} \rangle_{MC}}. \quad (28)$$

The first step in the derivation is to rewrite the partition function as a time ordered exponential using some *interaction representation*. We split the Hamiltonian into two parts, $H = H_1 + H_2$ and define the time dependent operators in the interaction picture as $O(\tau) = e^{\tau H_1} O e^{-\tau H_1}$. In a second step, the time ordered exponential is expanded into a power series,

$$\begin{aligned} Z &= Tr \left[e^{-\beta H_1} T e^{-\int_0^\beta d\tau H_2(\tau)} \right] \\ &= \sum_{n=0}^{\infty} \int_0^\beta d\tau_1 \dots \int_{\tau_{n-1}}^\beta d\tau_n Tr \left[e^{-(\beta-\tau_n)H_1} (-H_2) \dots e^{-(\tau_2-\tau_1)H_1} (-H_2) e^{-\tau_1 H_1} \right], \end{aligned} \quad (29)$$

which is a representation of the partition function of the form (26), namely the sum of all configurations $c = \{\tau_1, \dots, \tau_n\}$, $n = 0, 1, \dots$, $\tau_i \in [0, \beta)$ with weight

$$w_c = Tr \left[e^{-(\beta-\tau_n)H_1} (-H_2) \dots e^{-(\tau_2-\tau_1)H_1} (-H_2) e^{-\tau_1 H_1} \right] d\tau^n. \quad (30)$$

The *weak-coupling* continuous-time Monte Carlo approach is based on an expansion of Z in powers of the interaction U , and on an interaction representation in which the time evolution is determined by the *quadratic* part $H_0 + H_{\text{bath}} + H_{\text{mix}}$ of the Hamiltonian. The complementary “*strong-coupling*” approach is based on an expansion of Z in powers of the impurity-bath hybridization V , and an interaction representation in which the time evolution is determined by the *local* part $H_0 + H_U + H_{\text{bath}}$ of the Hamiltonian.

IV. WEAK-COUPPLING APPROACH

The weak-coupling continuous time impurity solver employs an expansion of the partition function in powers of $H_2 = H_U$. Equation (30) thus gives the weight of a configuration of n *interaction vertices*. Since $H_1 = H - H_2 = H_0 + H_{\text{bath}} + H_{\text{mix}}$ is quadratic, we can use Wick's theorem to evaluate the trace. The result is a product of two determinants of $n \times n$ matrices (one for each spin), whose elements are bath Green functions G_0 evaluated at the time intervals defined by the vertex positions:

$$\begin{aligned} \frac{w_c}{Z_0} &= (-U d\tau)^n \frac{1}{Z_0} \text{Tr} \left[e^{-(\beta-\tau_n)H_1} n_\uparrow n_\downarrow \dots e^{-(\tau_2-\tau_1)H_1} n_\uparrow n_\downarrow e^{-\tau_1 H_1} \right] \\ &= (-U d\tau)^n \prod_{\sigma} \det M_{\sigma}^{-1}, \end{aligned} \quad (31)$$

$$(M_{\sigma}^{-1})_{ij} = G_{0,\sigma}(\tau_i - \tau_j), \quad (32)$$

with $Z_0 = \text{Tr}[e^{-\beta H_1}]$ the partition function of the noninteracting model.

At this point, we encounter a problem. In the paramagnetic phase, where $G_{0,\uparrow} = G_{0,\downarrow}$, the product of determinants is positive, which means that for repulsive interaction ($U > 0$), odd perturbation orders yield negative weights. Except in the particle-hole symmetric case, where one can show that odd perturbation orders vanish, this will result in a severe sign problem. Fortunately, we can solve this sign problem by shifting the chemical potentials for up and down spins in an appropriate way. We rewrite the interaction term as

$$H_U = \frac{U}{2} \sum_{s,\sigma} (n_{\sigma} - \alpha_{\sigma}(s)) + \frac{U}{2} (n_{\uparrow} + n_{\downarrow}) - \frac{U}{4}, \quad (33)$$

$$\alpha_{\sigma}(s) = 1/2 + \sigma s(1/2 + \delta). \quad (34)$$

Here δ is some constant and $s = \pm 1$ an Ising variable. The constant $-U/4$ in Eq. (33) is irrelevant, while the contribution $U(n_{\uparrow} + n_{\downarrow})/2$ can be absorbed into the noninteracting Green function by shifting the chemical potential as $\mu \rightarrow \mu - U/2$. Explicitly, we redefine the bath Green function as $G_{0,\sigma}^{-1} = i\omega_n + \mu - \Delta_{\sigma} \rightarrow \tilde{G}_{0,\sigma}^{-1} = i\omega_n + \mu - U/2 - \Delta_{\sigma}$.

The introduction of an Ising variable s_i at each vertex position τ_i enlarges the configuration space exponentially. A configuration c now corresponds to a collection of Ising spin variables on the imaginary time interval: $c = \{(\tau_1, s_1), (\tau_2, s_2), \dots, (\tau_n, s_n)\}$. The weight of

these configurations are

$$\frac{w_c}{Z_0} = (-Ud\tau/2)^n \prod_{\sigma} \det \tilde{M}_{\sigma}^{-1}, \quad (35)$$

$$(\tilde{M}_{\sigma}^{-1})_{ij} = \tilde{G}_{0,\sigma}(\tau_i - \tau_j) - \alpha_{\sigma}(s_i)\delta_{ij}. \quad (36)$$

The Ising variables are in fact not needed to cure the sign problem. They have been introduced to symmetrize the interaction term and prevent ergodicity problems.

A. Sampling procedure and detailed balance

For ergodicity it is sufficient to insert/remove spins with random orientation at random times, because this allows in principle to generate all possible configurations. Furthermore, the random walk in configuration space must satisfy the detailed balance condition (27). Splitting the probability to move from configuration c_i to configuration c_j into a probability to *propose* the move and a probability to *accept* it,

$$p(c_i \rightarrow c_j) = p^{\text{prop}}(c_i \rightarrow c_j)p^{\text{acc}}(c_i \rightarrow c_j), \quad (37)$$

we arrive at the condition

$$\frac{p^{\text{acc}}(c_i \rightarrow c_j)}{p^{\text{acc}}(c_j \rightarrow c_i)} = \frac{p^{\text{prop}}(c_j \rightarrow c_i) |w(c_j)|}{p^{\text{prop}}(c_i \rightarrow c_j) |w(c_i)|}. \quad (38)$$

There is some flexibility in choosing the proposal probabilities. A reasonable choice for the insertion/removal of a spin is the following (illustrated in Fig. 3):

- *Insertion*

Pick a random time in $[0, \beta)$ and a random direction for the new spin:

$$p^{\text{prop}}(n \rightarrow n+1) = (1/2)(d\tau/\beta),$$

- *Removal*

Pick a random spin: $p^{\text{prop}}(n+1 \rightarrow n) = 1/(n+1)$.

For this choice, the ratio of acceptance probabilities becomes

$$\frac{p^{\text{acc}}(n \rightarrow n+1)}{p^{\text{acc}}(n+1 \rightarrow n)} = \frac{\beta U}{n+1} \prod_{\sigma=\uparrow,\downarrow} \frac{|\det(M_{\sigma}^{(n+1)})^{-1}|}{|\det(M_{\sigma}^{(n)})^{-1}|}, \quad (39)$$

and the random walk can thus be implemented for example on the basis of the Metropolis algorithm, *i.e.* the proposed move from n to $n \pm 1$ is accepted with probability $\min[1, p^{\text{acc}}(n \rightarrow n \pm 1)/p^{\text{acc}}(n \pm 1 \rightarrow n)]$.

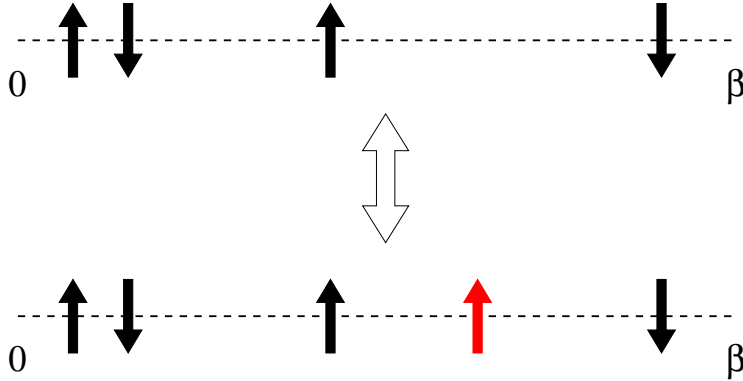


FIG. 3: Local update in the continuous-time auxiliary field method. The dashed line represents the imaginary time interval $[0, \beta)$. We increase the perturbation order by adding a spin with random orientation at a random time. The perturbation order is decreased by removing a randomly chosen spin.

B. Determinant ratios and fast matrix updates

From Eq. (39) it follows that each update requires the calculation of a ratio of two determinants. Computing the determinant of a matrix of size $(n \times n)$ is an $O(n^3)$ operation. However, each insertion or removal of a spin merely changes one row and one column of the matrix M_σ^{-1} . We will now show that it is therefore possible to evaluate the ratio in Eq. (39) in a time $O(n^2)$ (insertion) or $O(1)$ (removal).

The objects which are stored and manipulated during the simulation are, besides the lists of the times $\{\tau_i\}$ and spins $\{s_i\}$, the matrices $M_\sigma = (G_{0\sigma})^{-1}$. Inserting a spin adds a new row and column to M_σ^{-1} . We define the blocks (omitting the σ index)

$$(M^{(n+1)})^{-1} = \begin{pmatrix} (M^{(n)})^{-1} & Q \\ R & S \end{pmatrix}, \quad M^{(n+1)} = \begin{pmatrix} \tilde{P} & \tilde{Q} \\ \tilde{R} & \tilde{S} \end{pmatrix}, \quad (40)$$

where Q , R , S denote $(n \times 1)$, $(1 \times n)$, and (1×1) matrices, respectively, which contain the contribution of the added spin. The determinant ratio needed for the acceptance/rejection probability is then given by

$$\frac{\det(M^{(n+1)})^{-1}}{\det(M^{(n)})^{-1}} = \frac{1}{\det \tilde{S}} = S - [R][M^{(n)}Q]. \quad (41)$$

As we store $M^{(n)}$, computing the acceptance/rejection probability of an insertion move is an $O(n^2)$ operation. If the move is accepted, the new matrix $M^{(n+1)}$ is computed out of

$M^{(n)}$, Q , R , and S , also in a time $O(n^2)$:

$$\tilde{S} = (S - [R][M^{(n)}Q])^{-1}, \quad (42)$$

$$\tilde{Q} = -[M^{(n)}Q]\tilde{S}, \quad (43)$$

$$\tilde{R} = -\tilde{S}[RM^{(n)}], \quad (44)$$

$$\tilde{P} = N^{(n)} + [M^{(n)}Q]\tilde{S}[RM^{(n)}]. \quad (45)$$

It follows from Eq. (41) that the calculation of the determinant ratio for removing a spin is $O(1)$, since it is just element \tilde{S} , and from the above formulas we also immediately find the elements of the reduced matrix:

$$M^{(n)} = \tilde{P} - \frac{[\tilde{Q}][\tilde{R}]}{\tilde{S}}. \quad (46)$$

C. Measurement of the Green function

To compute the contribution of a configuration c to the Green function, $G_\sigma^c(\tau)$, we insert in Eq. (??) a creation operator d^\dagger at time 0 and an annihilation operator d at time τ and divide by w_c . Wick's theorem then leads to the expression

$$G_\sigma^c(\tau) = G_{0\sigma}(i\omega_n) - \sum_k G_{0\sigma}(\tau - \tau_k) \sum_l [M_\sigma]_{kl} G_{0\sigma}(\tau_l). \quad (47)$$

The impurity Green function is obtained as $G(\tau) = \langle G_\sigma^c(\tau) \rangle_{MC}$.

It is possible to accumulate the Fourier components of the Green function directly. Using translational invariance of the Green functions one finds

$$G_\sigma^c(i\omega_n) = G_{0\sigma}(i\omega_n) - G_{0\sigma}(i\omega_n) \sum_{k,l} \frac{1}{\beta} e^{i\omega_n(\tau_k - \tau_l)} [M_\sigma]_{kl} G_{0\sigma}(i\omega_n), \quad (48)$$

so that $G(i\omega_n) = G_0(i\omega_n) - \frac{1}{\beta} (G_0(i\omega_n))^2 \langle \sum_{k,l} e^{i\omega_n(\tau_k - \tau_l)} [M_\sigma]_{kl} \rangle_{MC}$. Note that because the bath Green function has the high-frequency behavior $G_0(i\omega_n) \sim 1/i\omega_n$, the impurity Green function will inherit the correct high-frequency tail.

D. Generalization - multiorbital and cluster impurity problems

The generalization of the weak-coupling method to impurity clusters is straight forward. All we have to do is to add a site index to the interaction vertices (or auxiliary Ising spin

variables) and sample the vertices (spins) on a family of n_{sites} imaginary time intervals. In principle, the weak-coupling solver can also be used to simulate lattice models, since the only difference to a multi-site impurity problem is the definition of the bath G_0 . However, the $O(\beta^3)$ scaling is not competitive with the BSS algorithm.

General four fermion terms as in (2) are, at least in principle, also easily dealt with. One simply expands the partition function in powers of all the interaction terms U^{abcd} . The trace over the impurity and bath degrees of freedom then again yields a determinant of a matrix whose size is equal to the total perturbation order. To reduce the sign problem, it is advantageous to introduce auxiliary fields α and replace

$$\sum_{abcd} U^{abcd} d_a^\dagger d_b^\dagger d_c d_d \rightarrow - \sum_{abcd} U^{abcd} (d_a^\dagger d_c - \alpha_{ac})(d_b^\dagger d_d - \alpha_{bd}), \quad (49)$$

with an appropriate shift in the quadratic part of the Hamiltonian. However, in general, it will not be possible to completely eliminate the sign problem by a suitable choice of the parameters α . Furthermore, since the number of interaction terms grows like $O(n_{\text{orbital}}^4)$ the computational cost rapidly escalates. In practice, the strong coupling approach discussed in the following section turns out to be a more suitable approach for single-site multi-orbital problems with complex interaction terms.

V. STRONG-COUPLING APPROACH - EXPANSION IN THE IMPURITY-BATH HYBRIDIZATION

A continuous-time Monte Carlo method, which is in many ways complementary to the weak-coupling approach, is based on an expansion of the partition function in powers of the impurity-bath hybridization V . Here, we decompose the Hamiltonian as $H_2 = H_{\text{mix}}$ and $H_1 = H - H_2 = H_0 + H_U + H_{\text{bath}}$. Since $H_2 \equiv H_2^{d^\dagger} + H_2^d = \sum_{\sigma,p} V_p^\sigma d_\sigma^\dagger a_{p,\sigma} + \sum_{\sigma,p'} V_{p'}^{\sigma*} d_\sigma a_{p',\sigma}^\dagger$ has two terms, corresponding to electrons hopping from the bath to the impurity and from the impurity back to the bath, only even perturbation orders contribute to Eq. (29). Furthermore, at perturbation order $2n$ only the $(2n)!/(n!)^2$ terms corresponding to n creation operators d^\dagger and n annihilation operators d will contribute. We can therefore write the

partition function as a sum over configurations $c = \{\tau_1, \dots, \tau_n; \tau'_1, \dots, \tau'_n\}$:

$$Z = \sum_{n=0}^{\infty} \int_0^{\beta} d\tau_1 \dots \int_{\tau_{n-1}}^{\beta} d\tau_n \int_0^{\beta} d\tau'_1 \dots \int_{\tau'_{n-1}}^{\beta} d\tau'_n \\ \times \text{Tr} \left[e^{-\beta H_1} T H_2^d(\tau_n) H_2^{d\dagger}(\tau'_n) \dots H_2^d(\tau_1) H_2^{d\dagger}(\tau'_1) \right]. \quad (50)$$

Since the time evolution of the AIM (given by H_1) does not rotate the spin, there is an additional constraint, namely that the configuration contains an equal number of creation and annihilation operators for both for spin up and spin down. Taking this into account and writing out the expressions for H_2^d and $H_2^{d\dagger}$ explicitly, we find

$$Z = Z_{\text{bath}} \sum_{\{n_{\sigma}\}} \prod_{\sigma} \int_0^{\beta} d\tau_1^{\sigma} \dots \int_{\tau_{n_{\sigma}-1}^{\sigma}}^{\beta} d\tau_{n_{\sigma}}^{\sigma} \int_0^{\beta} d\tau'_1{}^{\sigma} \dots \int_{\tau'_{n_{\sigma}-1}{}^{\sigma}}^{\beta} d\tau'_{n_{\sigma}}{}^{\sigma} \\ \times \text{Tr}_d \left[e^{-\beta H_{\text{loc}}} T \prod_{\sigma} d_{\sigma}(\tau_{n_{\sigma}}^{\sigma}) d_{\sigma}^{\dagger}(\tau'_{n_{\sigma}}{}^{\sigma}) \dots d_{\sigma}(\tau_1^{\sigma}) d_{\sigma}^{\dagger}(\tau'_1{}^{\sigma}) \right] \\ \times \frac{1}{Z_{\text{bath}}} \text{Tr}_a \left[e^{-\beta H_{\text{bath}}} T \prod_{\sigma} \sum_{p_1, \dots, p_{n_{\sigma}}} \sum_{p'_1, \dots, p'_{n_{\sigma}}} V_{p_1}^{\sigma} V_{p'_1}^{\sigma*} \dots V_{p_{n_{\sigma}}}^{\sigma} V_{p'_{n_{\sigma}}}^{\sigma*} \\ a_{\sigma, p_{n_{\sigma}}}^{\dagger}(\tau_{n_{\sigma}}^{\sigma}) a_{\sigma, p'_{n_{\sigma}}}(\tau'_{n_{\sigma}}{}^{\sigma}) \dots a_{\sigma, p_1}^{\dagger}(\tau_1^{\sigma}) a_{\sigma, p'_1}(\tau'_1{}^{\sigma}) \right], \quad (51)$$

where we have used the fact that H_1 does not mix the impurity and the bath to separate the d and a operators. $Z_{\text{bath}} = \text{Tr}_a e^{-\beta H_{\text{bath}}}$, and $H_{\text{loc}} = H_0 + H_U$. Introducing the β -antiperiodic *hybridization function* (13), which in the time-domain reads

$$\Delta_{\sigma}(\tau) = \sum_p \frac{|V_p|^2}{e^{-\epsilon_p \beta} + 1} \begin{cases} -e^{-\epsilon_p \tau} & \tau > 0 \\ -e^{-\epsilon_p(\beta + \tau)} & \tau < 0 \end{cases}, \quad (52)$$

the trace over the bath states can be expressed as

$$\frac{1}{Z_{\text{bath}}} \text{Tr}_a \left[e^{-\beta H_{\text{bath}}} T \prod_{\sigma} \sum_{p_1, \dots, p_{n_{\sigma}}} \sum_{p'_1, \dots, p'_{n_{\sigma}}} V_{p_1}^{\sigma} V_{p'_1}^{\sigma*} \dots V_{p_{n_{\sigma}}}^{\sigma} V_{p'_{n_{\sigma}}}^{\sigma*} \\ a_{\sigma, p_{n_{\sigma}}}^{\dagger}(\tau_{n_{\sigma}}^{\sigma}) a_{\sigma, p'_{n_{\sigma}}}(\tau'_{n_{\sigma}}{}^{\sigma}) \dots a_{\sigma, p_1}^{\dagger}(\tau_1^{\sigma}) a_{\sigma, p'_1}(\tau'_1{}^{\sigma}) \right] = \prod_{\sigma} \det M_{\sigma}^{-1}, \quad (53)$$

where M_{σ}^{-1} is the $(n_{\sigma} \times n_{\sigma})$ matrix with elements

$$M_{\sigma}^{-1}(i, j) = \Delta_{\sigma}(\tau'_i{}^{\sigma} - \tau_j^{\sigma}). \quad (54)$$

In the hybridization expansion method, the configuration space consists of all sequences $c = \{\tau_1^{\uparrow}, \dots, \tau_{n_{\uparrow}}^{\uparrow}; \tau'_1{}^{\uparrow}, \dots, \tau'_{n_{\uparrow}}{}^{\uparrow} | \tau_1^{\downarrow}, \dots, \tau_{n_{\downarrow}}^{\downarrow}; \tau'_1{}^{\downarrow}, \dots, \tau'_{n_{\downarrow}}{}^{\downarrow}\}$, of n_{\uparrow} creation and annihilation operators

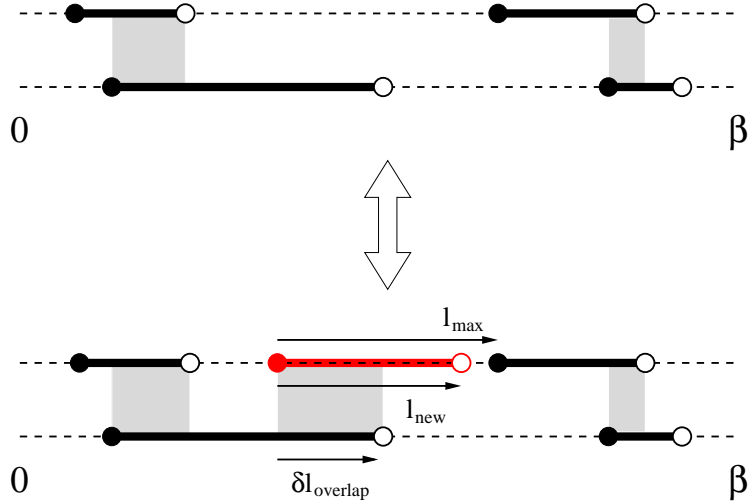


FIG. 4: Local update in the “segment” picture. The two segment configurations correspond to spin up and spin down. Each segment depicts a time interval in which an electron of the corresponding spin resides on the impurity (the end points are the locations of the operators d^\dagger and d). We increase the perturbation order by adding a segment or anti-segment of random length for random spin. The perturbation order is decreased by removing a randomly chosen segment.

for spin up ($n_\uparrow = 0, 1, \dots$), and n_\downarrow creation and annihilation operators for spin down ($n_\downarrow = 0, 1, \dots$). The weight of this configuration is

$$w_c = Z_{\text{bath}} \text{Tr}_d \left[e^{-\beta H_{\text{loc}}} T \prod_{\sigma} d_{\sigma}(\tau_{n_{\sigma}}^{\sigma}) d_{\sigma}^{\dagger}(\tau_{n_{\sigma}}^{\prime\sigma}) \dots d_{\sigma}(\tau_1^{\sigma}) d_{\sigma}^{\dagger}(\tau_1^{\prime\sigma}) \right] \times \prod_{\sigma} \det M_{\sigma}^{-1}(\tau_1^{\sigma}, \dots, \tau_{n_{\sigma}}^{\sigma}; \tau_1^{\prime\sigma}, \dots, \tau_{n_{\sigma}}^{\prime\sigma}) (d\tau)^{2n_{\sigma}}. \quad (55)$$

The trace factor represents the contribution of the impurity, which fluctuates between different quantum states, as electrons hop in and out. The determinants resum all the bath evolutions which are compatible with the given sequence of transitions (see Section ??).

To evaluate the trace factor, one may use the eigenbasis of H_{loc} . In this basis, the time evolution operator $e^{-\tau H_{\text{loc}}}$ is diagonal while the operators d_{σ} and d_{σ}^{\dagger} will produce transitions between eigenstates with amplitude ± 1 .

Because the time evolution does not flip the spin, the creation and annihilation operators for given spin have to alternate. This allows us to separate the operators for spin up from those for spin down and to depict the time evolution by a *collection of segments* (each segment representing a time interval in which an electron of spin up or down resides on

the impurity). At each time, the eigenstate of the impurity follows immediately from the segment representation and we can easily compute the trace factor as

$$\text{Tr}_d \left[e^{-\beta H_{\text{loc}}} T \prod_{\sigma} d_{\sigma}(\tau_{n_{\sigma}}^{\sigma}) d_{\sigma}^{\dagger}(\tau'_{n_{\sigma}}) \dots d_{\sigma}(\tau_1^{\sigma}) d_{\sigma}^{\dagger}(\tau'_1) \right] = s \exp \left[\mu(l_{\uparrow} + l_{\downarrow}) - U l_{\text{overlap}} \right], \quad (56)$$

with s a permutation sign, l_{σ} the total “length” of the segments for spin σ and l_{overlap} the total length of the overlap between up and down segments. The lower panel of Fig. 4 shows a configuration with 3 segments for spin up and two segments for spin down; the time intervals where segments overlap, indicated by gray rectangles, correspond to a doubly occupied impurity and cost a repulsion energy U .

A. Sampling procedure and detailed balance

For ergodicity, it is sufficient to insert and remove pairs of creation and annihilation operators (segments or anti-segments) for spin up and down. One possible strategy for inserting a segment is the following: we pick a random time in $[0, \beta)$ for the creation operator. If it falls on an existing segment, the impurity is already occupied and the move is rejected. If it falls on an empty space, we compute l_{max} , the length from this position to the next segment (in the direction of increasing τ). If there are no segments, $l_{\text{max}} = \beta$. The position of the new annihilation operator is then chosen randomly in this interval of length l_{max} (see Fig. 4). If we propose to remove a randomly chosen segment for this spin, then the proposal probabilities are

$$p^{\text{prop}}(n_{\sigma} \rightarrow n_{\sigma} + 1) = \frac{d\tau}{\beta} \frac{d\tau}{l_{\text{max}}}, \quad p^{\text{prop}}(n_{\sigma} + 1 \rightarrow n_{\sigma}) = \frac{1}{n_{\sigma} + 1}, \quad (57)$$

and the ratio of acceptance probabilities therefore becomes

$$\frac{p^{\text{acc}}(n_{\sigma} \rightarrow n_{\sigma} + 1)}{p^{\text{acc}}(n_{\sigma} + 1 \rightarrow n_{\sigma})} = \frac{\beta l_{\text{max}}}{n_{\sigma} + 1} e^{\mu l_{\text{new}} - U \delta l_{\text{overlap}}} \frac{|\det(M_{\sigma}^{(n_{\sigma}+1)})^{-1}|}{|\det(M_{\sigma}^{(n_{\sigma})})^{-1}|}. \quad (58)$$

Here, l_{new} is the length of the new segment, and $\delta l_{\text{overlap}}$ the change in the overlap. Again, we compute the ratio of determinants using the fast update formulas discussed in Section IV B.

B. Measurement of the Green function

The strategy here is to create configurations which contribute to the Green function measurement by decoupling the bath from a given pair of creation and annihilation operators in c . The idea is to write

$$g(\tau) = \frac{1}{Z} \sum_c w_c^{d(\tau)d^\dagger(0)} = \frac{1}{Z} \sum_c w_c^{(\tau,0)} \frac{w_c^{d(\tau)d^\dagger(0)}}{w_c^{(\tau,0)}}, \quad (59)$$

where $w_c^{d(\tau)d^\dagger(0)}$ denotes the weight of configuration c with and additional operator $d^\dagger(0)$ and $d(\tau)$ in the trace factor, and $w_c^{(\tau,0)}$ the complete weight corresponding to the enlarged operator sequence (including enlarged hybridization determinants). Since the trace factors of both weights are identical,

$$\frac{w_c^{d(\tau)d^\dagger(0)}}{w_c^{(\tau,0)}} = \frac{\det M_c^{-1}}{\det(M_c^{(\tau,0)})^{-1}} = (M_c^{(\tau,0)})_{j,i}, \quad (60)$$

with i and j denoting the row and column corresponding to the new operators d^\dagger and d in the enlarged $(M_c^{(\tau,0)})^{-1}$. Hence, the measurement formula for the Green function becomes

$$G(\tau) = \frac{1}{Z} \sum_{\bar{c}} w_{\bar{c}} \sum_{i,j} \frac{1}{\beta} \Delta(\tau, \tau_i - \tau'_j) (M_{\bar{c}})_{ji} = \left\langle \sum_{i,j} \frac{1}{\beta} \Delta(\tau, \tau_i - \tau'_j) M_{ji} \right\rangle_{MC}, \quad (61)$$

with $\Delta(\tau, \tau') = \delta(\tau - \tau')$ for $\tau' > 0$, and $\Delta(\tau, \tau') = -\delta(\tau - \tau' - \beta)$ for $\tau' < 0$.

We may Fourier transform Eq. (61) to obtain a measurement formula for the Fourier coefficients of the Green function,

$$G(i\omega_n) = \left\langle \sum_{i,j} \frac{1}{\beta} e^{i\omega_n(\tau_i - \tau'_j)} M_{ji} \right\rangle_{MC}. \quad (62)$$

Note that in contrast to the weak-coupling approach, where the Green function is measured as a $O(1/(i\omega_n)^2)$ correction to the bath Green function, Eq. (62) does not automatically yield the correct high frequency tails. It is thus advantageous to accumulate the coefficients of an expansion of the Green function in Legendre polynomials.

C. Generalization - Matrix formalism

It is obvious from the derivation in Section V that the hybridization expansion formalism is applicable to general classes of impurity models. Because the trace factor in the weight

(55) is computed exactly, H_{loc} can contain essentially arbitrary interactions (e. g. spin-exchange terms in multi-orbital models), degrees of freedom (e. g. spins in Kondo-lattice models) or constraints (e. g. no double occupancy in the t - J model).

For multi-orbital impurity models with density-density interaction, the segment formalism is still applicable: we have now a collection of segments for each *flavor* α (orbital, spin) and the trace factor can still be computed from the length of the segments (chemical potential contribution) and the overlaps between segments of different flavor (interaction terms).

If H_{loc} is not diagonal in the occupation number basis defined by the d_{α}^{\dagger} , the calculation of $\text{Tr}_d[e^{-\beta H_{\text{loc}}} T \prod_{\alpha} d_{\alpha}(\tau_{n_{\alpha}}^{\alpha}) d_{\alpha}^{\dagger}(\tau_{n_{\alpha}}^{\prime\alpha}) \dots d_{\sigma}(\tau_1^{\alpha}) d_{\alpha}^{\dagger}(\tau_1^{\prime\alpha})]$ becomes more involved. We now have to compute the trace explicitly in some basis of H_{loc} – for example the eigenbasis, in which the time evolution operators $e^{-H_{\text{loc}}\tau}$ become diagonal. The operators d_{α} and d_{α}^{\dagger} are expressed as matrices in this eigenbasis, and the evaluation of the trace factor thus involves the multiplication of matrices whose size is equal the size of the Hilbert space of H_{loc} . Since the dimension of the Hilbert space grows *exponentially* with the number of flavors, the calculation of the trace factor becomes the computational bottleneck of the simulation, and the matrix formalism is therefore restricted to a relatively small number of flavors (up to about 10).

An important point is the use of *conserved quantum numbers* (typically particle number for spin up and spin down, momentum, ...). If the eigenstates of H_{loc} are grouped according to these quantum numbers, the operator matrices will acquire a sparse block structure, because for example $d_{\uparrow,q}^{\dagger}$ will connect the states corresponding to quantum numbers $m = \{n_{\uparrow}, n_{\downarrow}, K\}$ to those corresponding to $m' = \{n_{\uparrow} + 1, n_{\downarrow}, K + q\}$ (if they exist). Checking the compatibility of the operator sequence with a given starting block furthermore allows one to find the (potentially) contributing quantum number sectors without any matrix multiplications. The evaluation of the trace is thus reduced to a block matrix multiplication of the form

$$\sum_{\text{contr. } m} \text{Tr}_m \left[\dots (O)_{m'',m'}(e^{-(\tau'-\tau)H_{\text{loc}}})_{m'}(O)_{m',m}(e^{-\tau H_{\text{loc}}})_m \right], \quad (63)$$

where O is either a creation or annihilation operator, m denotes the index of the matrix block, and the sum runs over those sectors which are compatible with the operator sequence.