7 LDA+DMFT for Strongly Correlated Materials

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Contents

1   DFT and DMFT: Role of reference systems

In this lecture we give an introduction to the theoretical description of interacting electron system based on non-perturbative, strong-coupling expansions around optimal reference systems. The Density Functional Theory (DFT) and its Local Density Approximation (LDA) based on the simplest reference system related to the homogeneous electron gas with constant external potential with the same Coulomb electron-electron interactions (see Fig. 1). Such a reference system can be solved via the numerically exact diffusion Quantum Monte Carlo scheme for the ground state energy as function of electron density [1]. On other hand, the Dynamical Mean-Field Theory (DMFT) for strongly interacting fermionic systems is based on the strong coupling expansion around an effective impurity reference system (Fig. 1). This scheme become exact in the limit of infinite lattice dimension ($z \to \infty$) [2].

For finite lattice dimension we can start from the DMFT reference system and use analytical perturbation theory for non-local correlation effects. The frequency dependent effective impurity DMFT problem can nowadays be efficiently solved within the continuous-time Quantum Monte Carlo (ct-QMC) scheme [3]. Therefore the perturbation theory needs to be formulated in the action path integral formalism. We give a brief introduction to the path integral over fermionic Grassmann fields and formulate a general scheme for the expansion around the DMFT solution using a special dual space transformation. We discuss here a general way to include nonlocal correlations beyond DMFT [4] which based on the dual-fermion path-integral formalism [5].

Consider the noninteracting, "kinetic" part H_t of the Hubbard model first [6]. This is fixed by specifying the hopping-matrix elements t_{ij} between sites i and j. In the absence of the local Hubbard-interaction term, H_t is easily diagonalized. For a Hubbard model on a lattice, diagonalization is achieved by Fourier transformation of hopping parameters to $\mathbf{k}$-space, and one has the normal "band structure". For a single orbital model it is particularly simple: $\varepsilon_{\mathbf{k}} = t_{\mathbf{k}}$ with band width W. If, on the other hand, only the local part of the Hamiltonian is kept, i.e., the Hubbard interaction H_U with interaction strength U and the local term of H_t fixed by the on-site energy ε_0, the diagonalization of the Hamiltonian is trivial again and reduces to the diagonalization of a single "Hubbard atom".

The great success of the DMFT approach is related with the interpolation between these two limits [7]. For the half-filled Hubbard model on the infinite-dimension Bethe lattice at half-filling the DMFT gives the exact description of the Mott-transition [8] between the weak-coupling ($U/W \ll 1$) metallic state and the strong-coupling ($U/W \gg 1$) insulating paramagnetic state [9]. In a nutshell, DMFT maps the correlated Hubbard lattice problem onto the self-consistent solution of an effective Anderson impurity problem with a single interacting Hubbard atom (interaction strength U) in a non-interacting fermionic bath (which mimics the rest of the crystal).

Now we can think how to incorporate nonlocal correlations beyond the DMFT: since the Hubbard and the Anderson-impurity model share the same interaction part, one can think of the Hubbard model as the impurity model plus a residual term $\propto (t_{\mathbf{k}} - \Delta_\nu)$ and treat this *perturbatively*. Since this term is frequency dependent we need a novel perturbation theory based on the

Lattice problem   Reference system
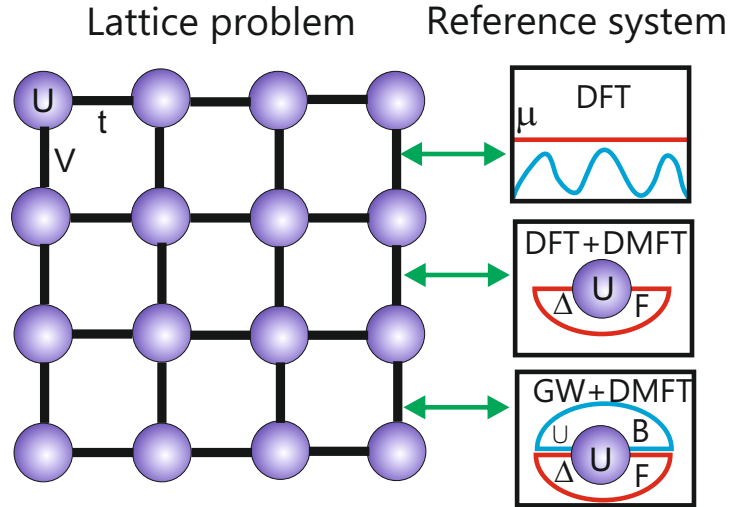
Fig. 1: *Schematic representation of reference systems in many-body approaches to lattice-fermion models: (i) Density-functional theory (DFT) with the interacting homogeneous electron gas as a reference system, defined by a constant external potential μ. (ii) Dynamical mean-field theory (DMFT) with an effective impurity problem as a reference system, defined by a fermionic bath, specified by a hybridization function Δ. (iii) GW+DMFT with correlated atom in a fermionic (Δ) and a bosonic bath (Λ) due to effects of the frequency-dependent screening of long-range Coulomb (V) interactions.*

action formalism. One may view this idea as a generalization of the Kohn-Sham idea in density functional theory (DFT) [10, 11] of an optimal reference system, but with a crucial difference. Here, not an interacting homogeneous electron gas, but an effective impurity model, tailored to the problem of strong correlations, serves as the reference system, see Fig. 1. Since at the zeroth order of this perturbative expansion, i.e., on the level of the DMFT problem, we already have an interacting problem and since the perturbation is momentum and frequency dependent, one is forced to replace the Hamiltonians by actions within the path-integral formalism. Note that the fermion path integral can also be used to formulate the DMFT itself [7, 13]. Now, the separation of the local and nonlocal terms is achieved by a Hubbard-Stratonovich transformation applied to the single-particle $(t_{\mathbf{k}} - \Delta_\nu)$-term [5]. This provides us with a new action. Moreover, it is formally possible to integrate out the original local degrees of freedom and in this way generate an effective action in the transformed, so-called dual-fermion representation [5]. Note that integrating out the local degrees of freedom is not only a formal step but can be achieved in practice, by solving the impurity problem within the numerically exact ct-QMC method.

The dual action consists of a bare dual propagator (non-local part of the DMFT Green function) $\tilde{G}^0_{\mathbf{k},\nu}$ and a local but frequency-dependent effective potential related to scattering processes of two, three, and more dual particles on the impurity site. The simplest two-particle dual potential coincides with the fully connected part of the screened impurity interaction vertex $\gamma^\omega_{\nu\nu'}$, which can be calculated with the impurity ct-QMC solver as a function of bosonic (ω) and fermionic (ν, ν') Matsubara frequencies. Normally, correlations between three particles on the DMFT impurity site are much weaker than two-particle correlations and can be ignored. The same ap-

plies to higher-order terms. One can think of the dual-fermion formalism as an expansion in the order of local multi-particle correlation functions. This means that "bare" interactions between dual fermions are related with the connected part of the screened impurity vertex. Standard diagrammatic techniques can be applied for calculations of the bold dual propagator $\tilde{G}_{\mathbf{k},\nu}$, which allows to obtain the nonlocal self-energy for the original fermions [5] and to describe nonlocal correlations beyond the DMFT.

The dual-fermion approach is not necessarily bound to a specific starting point. However, the DMFT starting point is very efficient. Namely, it corresponds to the elimination of all local diagrams for any n-particle correlation of dual fermions when using the DMFT self-consistency equation, Eq. (46). In the dual space, this simply reduces to $\sum_{\mathbf{k}} \tilde{G}^0_{\mathbf{k},\nu} = 0$ and means that, on average over the whole Brillouin zone, Δ_ν optimally approximates the electron spectrum $\varepsilon_{\mathbf{k}}$, including its local correlation effects. Therefore, the noninteracting dual fermions correspond to strongly correlated DMFT quasiparticles, and the remaining nonlocal effects can be quite small and reasonably described by perturbative summations of dual diagrams. This also explains the notion "dual fermions".

2   Functional approach

We introduce a general functional approach which will cover the DFT, Dynamical Mean Field Theory (DMFT) and Baym-Kadanoff (BK) theories [12, 13]. Let us start from the full many–body Hamiltonian describing electrons moving in the periodic external potential of ions $V(\mathbf{r})$ with the chemical potential μ and interacting via Coulomb law: $U(\mathbf{r}-\mathbf{r}') = 1/|\mathbf{r}-\mathbf{r}'|$. We use the atomic units $\hbar = m = e = 1$. In the field-operator representation, the Hamiltonian has the form

$$H = \sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r}) \left(-\frac{\nabla^2}{2} + V(\mathbf{r}) - \mu \right) \hat{\psi}_\sigma(\mathbf{r}) + \frac{1}{2} \sum_{\sigma\sigma'} \int d\mathbf{r} \int d\mathbf{r}' \hat{\psi}^\dagger_\sigma(\mathbf{r}) \hat{\psi}^\dagger_{\sigma'}(\mathbf{r}')\, U(\mathbf{r}-\mathbf{r}')\, \hat{\psi}_{\sigma'}(\mathbf{r}') \hat{\psi}_\sigma(\mathbf{r})$$

(1)

We can always use the single-particle orthonormal basis set in solids $\varphi_n(\mathbf{r})$ for example Wannier orbitals with full set of quantum numbers, e.g., site, orbital and spin index: $k = (i, m, \sigma)$ and expand the fields in creation and annihilation operators

$$\hat{\psi}(\mathbf{r}) = \sum_n \varphi_n(\mathbf{r})\, \hat{c}_n \qquad \hat{\psi}^\dagger(\mathbf{r}) = \sum_n \varphi_n^*(\mathbf{r})\, \hat{c}_n^\dagger.$$

(2)

Going from fermionic operators to the Grassmann variables $\{c_k^*, c_k\}$ we can write the functional integral representation for partition function of the many-body Hamiltonian in the imaginary time domain using the Euclidean action S

$$Z = \int \mathcal{D}[c^*, c]\, e^{-S}, \qquad S = \sum_{12} c_1^*(\partial_\tau + t_{12})c_2 + \frac{1}{2} \sum_{1234} c_1^* c_2^*\, U_{1234}\, c_4 c_3,$$

(3)

where the one- and two-electron matrix elements are defined as

$$t_{12} = \int d\mathbf{r} \, \varphi_1^*(\mathbf{r}) \left(-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \mu \right) \varphi_2(\mathbf{r}) \tag{4}$$

$$U_{1234} = \int d\mathbf{r} \int d\mathbf{r}' \, \varphi_1^*(\mathbf{r})\varphi_2^*(\mathbf{r}') \, U(\mathbf{r}-\mathbf{r}') \, \varphi_3(\mathbf{r})\varphi_4(\mathbf{r}').$$

and we use the following short definition of the sum:

$$\sum_1 \cdots \equiv \sum_{im} \int d\tau \cdots \tag{5}$$

The one-electron Green function defined via a simplest non-zero correlation function for fermions [14] can be written in the path-integral formalism [15–17] as

$$G_{12} = -\left\langle c_1 c_2^* \right\rangle_S = -\frac{1}{Z} \int \mathcal{D}[c^*, c] \, c_1 c_2^* \, \exp(-S). \tag{6}$$

The Baym-Kadanoff functional [12] gives the one–particle Green function and the total free energy at its stationary point. In order to construct the exact functional of the Green function (Baym-Kadanoff) we modify the action by introducing the source term J

$$S[J] = S + \sum_{12} c_1^* J_{12} \, c_2. \tag{7}$$

The partition function Z, or equivalently the free energy of the system F, becomes a functional of the auxiliary source field

$$Z[J] = e^{-F[J]} = \int \mathcal{D}[c^*, c] \, e^{-S'[J]}. \tag{8}$$

Variation of this source function gives all correlation functions, for example the Green function

$$G_{12} = \left. \frac{\delta F[J]}{\delta J_{21}} \right|_{J=0}. \tag{9}$$

The Baym-Kadanoff functional Γ is obtained by the Legendre transform from variable J to G

$$\Gamma[G] = F[J] - \mathrm{Tr} \, JG, \tag{10}$$

This is a non-trivial step and uses the possibility of inverting the Eq. (9) for arbitrary J and find $J = J(G)$ which may be multivalued for strongly correlated systems [18].

We can use the standard decomposition of the free energy functional Γ into the single particle part and the correlated part

$$\Gamma[G] = \mathrm{Tr} \ln G - \mathrm{Tr} \, \Sigma G + \Phi[G], \tag{11}$$

were Σ is single particle self-energy and $\Phi[G]$ is a correlated part of the Baym-Kadanoff functional and is equal to the sum of all two-particle irreducible diagrams. At the stationary point this functional gives the free energy of the system. In practice, $\Phi[G]$ is not known for interacting electron systems, which is similar to the problem in the density functional theory. There is a formal expression for the "exchange-correlation" part of this functional via all connecting two-particle diagrams [13, 19, 20].

3　Density Functional Theory

The general functional approach reduces to density-functional theory, if one only uses the diagonal part in space-time of the Green function, which corresponds to the one-electron density

$$n(1) = G_{12}\,\delta_{12} = \langle c_1^* c_1 \rangle_S, \tag{12}$$

Consider a general interacting Hamiltonian with "λ-scaled" interaction

$$\hat{H} = \hat{T} + \lambda \hat{U}, \tag{13}$$

which depends on the coupling constant λ as a parameter. The same is true for the effective action functional [20]

$$\Gamma = \Gamma[n, \lambda]. \tag{14}$$

Clearly n, λ are to be considered as two independent variables. Note, however, that this does not prevent the exact expectation value n_g from depending on λ: this dependence is fixed by the variational principle

$$\left(\frac{\delta \Gamma[n, \lambda]}{\delta n} \right)_{n_g} = 0. \tag{15}$$

The functional $\Gamma[n, \lambda]$ is defined as

$$\Gamma[n, \lambda] = F[J, \lambda] - J(1)\, n(1), \tag{16}$$

where J is a functional of n and λ. This functional dependence is provided by the equation

$$\frac{\delta F[J, \lambda]}{\delta J(1)} = n(1).$$

We can find a formally exact expression for the $\Gamma[n]$ functional within the inversion method [19]. Let us expand all the quantities in Eqn (16) in terms of λ;

$$\begin{aligned}
J[n, \lambda] &= J_0[n] + \lambda J_1[n] + \lambda^2 J_2[n] + \dots, \\
F[J, \lambda] &= F_0[J] + \lambda F_1[J] + \lambda^2 F_2[J] + \dots, \\
\Gamma[n, \lambda] &= \Gamma_0[n] + \lambda \Gamma_1[n] + \lambda^2 \Gamma_2[n] + \dots.
\end{aligned} \tag{17}$$

Comparison of the two sides in Eqn (16) for different orders of λ,

$$\sum \lambda^i \Gamma_i[n] = \sum \lambda^i F_i \left(\sum \lambda^k J_k[n] \right) - \sum \lambda^i J_i(1)\, n(1), \tag{18}$$

leads to the formally exact expression for $\Gamma_i[n]$,

$$\Gamma_i[n] = F_i[J_0] + \sum_{k=1}^{i} \frac{\delta F_{i-k}[J_0]}{\delta J_0(1)} J_k(1) - J_i(1)\, n(1)$$

$$+ \sum_{m=2}^{i} \frac{1}{m!} \sum_{\substack{k_1+\dots+k_m \leq i \\ k_1,\dots,k_m \geq 1}} \frac{\delta^m F_{i-(k_1+\dots+k_m)}[J_0]}{\delta J_0(1)\dots\delta J_0(m)} J_{k_1}(1)\cdots J_{k_m}(m).$$

The functionals $\{F_i\,[J_0]\,\}$ and their derivatives are assumed to be known and obtained via a standard many-body Quantum Monte Carlo scheme. Since n and λ are considered to be independent, the functionals $J_i\,[n]$ can be obtained using,

$$\frac{\delta\Gamma_i\,[n]}{\delta n\,(1)} = -J_i\,(1)\,.\tag{19}$$

Of special importance for the DFT scheme is the zero-th order term

$$\Gamma_0\,[n] = F_0\,[J_0] - J_0\,(1)\,n\,(1)\,.\tag{20}$$

Using Eqn (19)

$$-J_0\,(1) = \frac{\delta F_0\,[J_0]}{\delta J_0\,(1')}\frac{\delta J_0\,(1')}{\delta n\,(1)} - J_0\,(1) - n\,(1')\frac{\delta J_0\,(1')}{\delta n\,(1)} \;\Rightarrow\; \left(\frac{\delta F_0\,[J_0]}{\delta J_0\,(1')} - n\,(1')\right)\frac{\delta J_0\,(1')}{\delta n\,(1)} = 0.$$

Strict convexity of $\Gamma_0\,[n]$ prohibits $(\delta J_0\,(1')\,/\delta n\,(1))$ from having zero eigenvalues. Thus we obtain that J_0 obeys the equation:

$$n\,(1) = \frac{\delta F_0\,[J_0]}{\delta J_0\,(1)}.\tag{21}$$

Hence J_0 is determined as a potential which generates the expectation value n in the *noninteracting* $(\lambda = 0)$ system. Notice that the same exact notion appears in the Kohn-Sham formalism [11]. We refer to this noninteracting system as Kohn-Sham (KS) system and J_0 as Kohn-Sham potential.

The Kohn–Sham potential for interacting system $V_{\mathrm{KS}} = V_{\mathrm{ext}} + V_H + V_{\mathrm{xc}}$ plays the role of the "constrained fields" J_i. Here V_{ext} is am external potential and V_H the Hartree potential. In principle, the exchange-correlation potential V_{xc} is known only for the homogeneous electron gas, therefore in all practical applications one uses a so-called local density approximation to DFT. In this case the DFT functional is defined in the following way

$$F_{\mathrm{DFT}}[n] = T_0[n] + V_{\mathrm{ext}}[n] + V_H[n] + V_{\mathrm{xc}}[n]\tag{22}$$

where T_0 is the kinetic energy of the non-interacting systems. Finally, if we define the total electron density and exchange density as
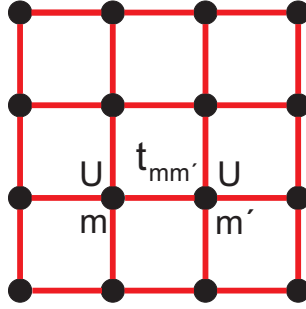
$$n(\mathbf{r}) = \sum_k \varphi_k^*(\mathbf{r})\varphi_k(\mathbf{r}) \quad\text{and}\quad n(\mathbf{r},\mathbf{r}') = \sum_k \varphi_k^*(\mathbf{r})\varphi_k(\mathbf{r}')\tag{23}$$

the DFT approach can be formulated as

$$T_0[n] + V_{\mathrm{ext}}[n] = \sum_k \int d\mathbf{r}\,\varphi_k^*(\mathbf{r})\left(-\frac{1}{2}\nabla^2 + V_{\mathrm{ext}}(\mathbf{r}) - \mu\right)\varphi_k(\mathbf{r})\tag{24}$$

$$V_H[n] = \frac{1}{2}\int d\mathbf{r}\,n(\mathbf{r})\,U(\mathbf{r}-\mathbf{r}')\,n(\mathbf{r}')\tag{25}$$

$$V_{\mathrm{xc}}[n] = -\frac{1}{2}\int d\mathbf{r}\,n(\mathbf{r},\mathbf{r}')\,U(\mathbf{r}-\mathbf{r}')\,n(\mathbf{r}',\mathbf{r}) + \sum_{i=2}^{\infty}\Gamma_i\,[n]\tag{26}$$

Fig. 2: *Generic Hubbard lattice for correlated lattice fermions with the local Coulomb interaction U and hopping parameters t: m can label different orbitals or lattice sites.*

and the local density approximation (LDA) to the DFT reads

$$V_{\mathrm{xc}}[n] = \int d\mathbf{r}\, n(\mathbf{r})\, \varepsilon_{\mathrm{xc}}\big(n(\mathbf{r})\big) \tag{27}$$

where $\varepsilon_{\mathrm{xc}}(n)$ is exchange correlation density for homogeneous electron Coulomb gas which can be calculated with the QMC scheme [1].

In the DFT scheme we lose information about the non-equal time Green function, which gives the single particle excitation spectrum as well as the k-dependence of the spectral function, and restrict ourself to only the ground-state energy of the many-electron system. Moreover, we lose information about all collective excitations in solids, such as plasmons or magnons, which can be obtained from the generalized susceptibility.

One of the most successful approaches to correlated materials is based on the combination of the DFT scheme with a Dynamical Mean Field Theory (DMFT) for strongly interacting d- or f-electrosn in the crystal. In the DMFT scheme one can obtain the numerically exact solution for the correlated part of the local functional. In the following section we discuss the general strong-coupling perturbation theory based on so-called dual fermion (DF) transformations [5] in the path integral formalism, which allows us to introduce the DMFT scheme as zero-order DF-expansion and shows a perturbative way to go beyond the DMFT approximations.

4 Dual Fermion approach with a general reference system

We start with a general lattice fermion model with the local Hubbard-like interaction vertex U. Generalization to the multi-orbital case is straightforward [21]. All equations will be written in matrix form, giving an idea of how to rewrite the dual fermion (DF) formula to the multi-orbital or multi-site case. The general strategy is related with the formally exact separation of the local and non-local correlation effects. We introduce auxiliary dual fermionic fields which will couple local correlated impurities or clusters back to the original lattice [5].

Using the path-integral formalism (Appendix A) the partition function of a general fermionic lattice system (Fig. 2) can be written in the following form as a functional integral over Grassmann variables $[c^*, c]$

$$Z = \int \mathcal{D}[c^*, c] \exp\big(-S_L[c^*, c]\big). \tag{28}$$
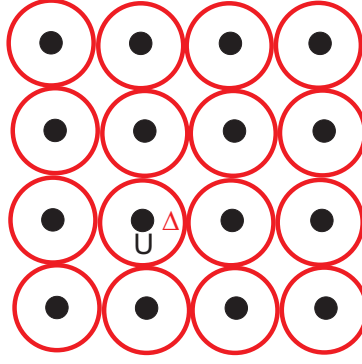
Fig. 3: *Schematic view on the real-space DMFT reference system.*

The original lattice action of interacting lattice fermions, similar to Eq. (3), can be written in Matsubara space as a sum of the lattice one-electron contributions with the Fourier transformed hopping $t_{\mathbf{k}}$ (or energy spectrum in the single-orbital case) and the local interaction part U

$$S_L[c^*, c] = -\sum_{\mathbf{k}\nu\sigma} c^*_{\mathbf{k}\nu\sigma}\big(i\nu + \mu - t_{\mathbf{k}}\big)\, c_{\mathbf{k}\nu\sigma} + \sum_i \int_0^\beta d\tau\, U\, n^*_{i\tau\uparrow} n_{i\tau\downarrow}\,. \tag{29}$$

In the following, $\nu = (2n{+}1)\pi/\beta, (\omega = 2n\pi/\beta), n = 0, \pm 1, \ldots$ are the fermionic (bosonic) Matsubara frequencies, β is the inverse temperature, $\tau \in [0, \beta)$ the imaginary time, and μ the chemical potential. The index i labels the lattice sites, m refers to different orbitals, σ is the spin projection and the $\mathbf{k}$-vectors are quasimomenta. In order to keep the notation simple, it is useful to introduce the combined index $|1\rangle \equiv |i, m, \sigma, \tau\rangle$ and assume summation over repeated indices. Translational invariance is assumed for simplicity in the following, although a real space formulation is straightforward. The local part of the action, S_U, may contain any type of local multi orbital interaction.

In order to formulate an expansion around the best possible reference action, Fig. 3, a quantum impurity (cluster) problem is introduced by a general frequency-dependent hybridization function Δ_ν and the same local interaction

$$S_\Delta[c^*_i, c_i] = -\sum_{\nu,\sigma} c^*_{i\nu\sigma}\big(i\nu + \mu - \Delta_\nu\big) c_{i\nu\sigma} + \sum_\nu U n^*_{i\nu\uparrow} n_{i\nu\downarrow}\,, \tag{30}$$

where Δ_ν is the effective hybridization matrix describing the coupling of the impurity to an auxiliary fermionic bath. The main motivation for rewriting the lattice action in terms of a quantum impurity model is that such a reference system can be solved numerically exactly for an arbitrary hybridization function using ct-QMC methods [3]. Using the locality of the hybridization function Δ_ν, the lattice action Eq. (29) can be rewritten exactly in terms of individual impurity models and the effective one-electron coupling $\Delta_\nu - t_{\mathbf{k}}$ between different impurities, Fig. 4,

$$S_L[c^*, c] = \sum_i S_\Delta[c^*_i, c_i] - \sum_{\mathbf{k}\nu\sigma} c^*_{\mathbf{k}\nu\sigma}\big(\Delta_\nu - t_{\mathbf{k}}\big) c_{\mathbf{k}\nu\sigma}\,. \tag{31}$$

We will find the condition for the optimal choice of the hybridization function later. Although we can solve the individual impurity model exactly, the effect of spatial correlations due to
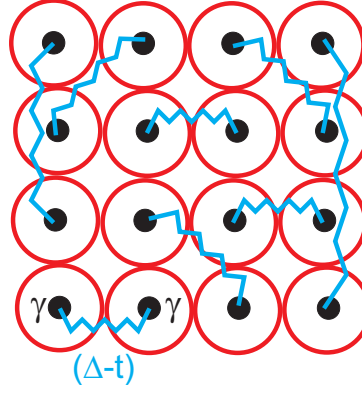
Fig. 4: *Schematic view on the non-local DF perturbation beyond a DMFT solution.*

the second term in Eq. (31) is very hard to treat, even perturbatively, since the impurity action is non-Gaussian and one cannot use of the Wick theorem. The main idea of the dual fermion transformation is the change of variables from strongly correlated fermions (c^*, c) to weakly correlated "dual" Grassmann fields (d^*, d) in the path-integral representation for the partition function of Eq. (3), followed by a simple perturbation treatment. The new variables were introduced through the following Hubbard-Stratonovich(HS)-transformation with the matrix $\widetilde{\Delta}_{\mathbf{k}\nu} = \Delta_\nu - t_{\mathbf{k}}$ in real space

$$e^{c_1^* \, \widetilde{\Delta}_{12} \, c_2} = \det \widetilde{\Delta} \int \mathcal{D}\left[d^*, d\right] e^{-d_1^* \widetilde{\Delta}_{12}^{-1} d_2 - d_1^* c_1 - c_1^* d_1}. \tag{32}$$

We can immediately seen that using this HS-transformation we "localize" the $[c_i^*, c_j]$ fermions: while on the left-hand side they are still "hopping" through the lattice, on the right-hand side they are localized on one site $[c_i^*, c_i]$.

With this reference system the lattice partition function becomes

$$\frac{Z}{Z_d} = \int \mathcal{D}[c^*, c, d^*, d] \exp\left(-S[c^*, c, d^*, d]\right) \tag{33}$$

with $Z_d = \det \widetilde{\Delta}$. The lattice action transforms to

$$S[c^*, c, d^*, d] = \sum_i S_\Delta^i + \sum_{\mathbf{k}, \nu, \sigma} d_{\mathbf{k}\nu\sigma}^* \left(\Delta_\nu - t_k\right)^{-1} d_{\mathbf{k}\nu\sigma}. \tag{34}$$

Hence the coupling between sites is transferred to a local coupling to the auxiliary fermions

$$S_\Delta^i[c_i^*, c_i, d_i^*, d_i] = S_\Delta[c_i^*, c_i] + \sum_{\nu, \sigma} \left(d_{i\nu\sigma}^* \, c_{i\nu\sigma} + c_{i\nu\sigma}^* \, d_{i\nu\sigma}\right) \tag{35}$$

For the last term we use the invariance of the trace so that the sum over all states labeled by $\mathbf{k}$ could be replaced by the equivalent summation over all sites by a change of basis in the second term. The crucial point is that the coupling to the auxiliary fermions is purely local and S_Δ^i decomposes into a sum of local terms. The lattice fermions can therefore be integrated out from S_Δ^i for each site i separately. This completes the change of variables

$$\frac{1}{Z_\Delta} \int \mathcal{D}[c^*, c] \exp\left(-S_\Delta^i[, c_i^*, c_i, d_i^* d_i]\right) = \exp\left(-\sum_{\nu\,\sigma} d_{i\nu\sigma}^* \, g_\nu d_{i\nu\sigma} - V_i[d_i^* d_i]\right), \tag{36}$$

where Z_Δ is the partition function of the impurity action Eq. (30) and g_ν is the exact impurity Green function

$$g_{12} = -\langle c_1 c_2^* \rangle_\Delta = \frac{1}{Z_\Delta} \int \mathcal{D}[c^*, c]\, c_1 c_2^*\, e^{-S_\Delta[c^*, c]}. \tag{37}$$

The above equation may be viewed as the defining equation for the dual potential $V[d^*, d]$. The choice of matrices in Eq. (32) ensures a particularly simple form of this potential. An explicit expression is found by expanding both sides of Eq. (36) and equating the resulting expressions order by order. Formally this can be done up to all orders and in this sense the transformation to the dual fermions is exact. For most applications, the dual potential is approximated by the first non-trivial interaction vertex

$$V[d^*, d] = \frac{1}{4} \sum_{1234} \gamma_{1234}\, d_1^* d_2^* d_4 d_3\,, \tag{38}$$

where for the local vertex the combined index $1 \equiv \{m\nu\sigma\}$ comprises orbital degrees of freedom (or cluster sites), frequency, and spin. γ is the exact, fully antisymmetric, reducible two-particle vertex of the local quantum impurity problem. With the present choice of normalization in the HS-transformation we did not "amputate" the impurity "legs" or g_{12} Greens function which will be a very useful choice for ct-QMC calculations of the local vertex for multi-orbital case. It is given then by the connected part of the local two-particle correlations function

$$\gamma_{1234} = \chi_{1234} - \chi_{1234}^0 \tag{39}$$

with the two-particle Green function of the local impurity (reference system) being defined as

$$\chi_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_\Delta = \frac{1}{Z_\Delta} \int \mathcal{D}[c^*, c]\, c_1 c_2 c_3^* c_4^*\, e^{-S_\Delta[c^*, c]}\,. \tag{40}$$

The disconnected part of a generalized susceptibility reads

$$\chi_{1234}^0 = g_{14} g_{23} - g_{13} g_{24}\,. \tag{41}$$

The single- and two-particle Green functions can be calculated using the ct-QMC Monte Carlo algorithms [3]. After integrating-out the lattice fermions, the dual action depends on the new variables only and for the one-orbital paramagnetic case reads

$$\tilde{S}[d^*, d] = -\sum_{\mathbf{k}\nu\sigma} d_{\mathbf{k}\nu\sigma}^*\, \tilde{G}_{0\mathbf{k}\nu}^{-1}\, d_{\mathbf{k}\nu\sigma} + \sum_i V_i[d_i^*, d_i] \tag{42}$$

while the bare dual Green function is has the form

$$\tilde{G}_{\mathbf{k}\nu}^0 = \left(\left(t_\mathbf{k} - \Delta_\nu \right)^{-1} - g_\nu \right)^{-1}. \tag{43}$$

This formula involves only the local Green function g_ν of the impurity model. It is important to note, that the HS-transformation to dual fermion variables, allows us to "perform the analytical amputation" of impurity "legs" which causes enormous problems in the multi-orbital ct-QMC
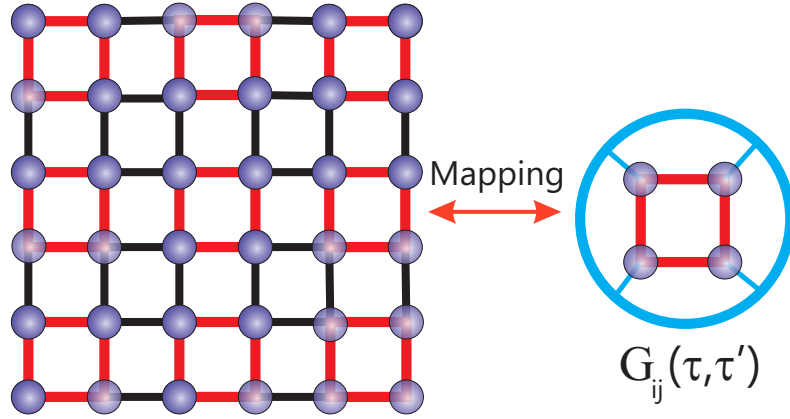
Fig. 5: *Schematic representation of a plaquette cluster-reference system for the square lattice.*

formalism. Transformation to the original DF-normalization where both dual G_d and real Green function have the same dimension unit reads

$$G_d = g\,\widetilde{G}\,g = G_{\text{DMFT}} - g \qquad G_{\text{DMFT}} = \left(g_\nu + \Delta_\nu - t_{\mathbf{k}}\right)^{-1}. \tag{44}$$

The Dual Fermion transformation allows us to use arbitrary reference systems and transform the strongly correlated lattice fermion problem to an effective action of weakly coupled dual quasiparticles. This is related with the fact that the bare dual Green function Eq. (44) related with the small non-local part of the DMFT lattice Green function and the main two-particle part of the bare interaction among dual fermions Eq. (38) is exactly equal to the fully screened impurity vertex of the reference system. The rest of the dual fermion problem is related with an optimal perturbation scheme for such an effective action Eq. (42).

5   Perturbation in Dual Space

For general multi-orbital multi-site dual fermion perturbation technique from arbitrary reference system (Fig. 5) we use the particle-hole notation for the local vertex and write the exact spin and imaginary time structure of the generalized connected susceptibility [5, 22]

$$\gamma^{\sigma\sigma'}_{1234}(\tau_1, \tau_2, \tau_3, \tau_4) = -\left\langle c_{1\sigma} c^*_{2\sigma} c_{3\sigma'} c^*_{4\sigma'} \right\rangle_\Delta + g^\sigma_{12} g^{\sigma'}_{34} - g^\sigma_{14} g^\sigma_{32}\, \delta_{\sigma\sigma'}.$$

Then the bare vertex of dual-fermion perturbation is related with the full impurity vertex, which in Matsubara space depends on two fermionic (ν, ν') and one bosonic (ω) frequencies. We also symmetrized vertex for charge density d- and spin s-channels

$$\gamma^{d/m}_{1234}(\nu, \nu', \omega) = \gamma^{\uparrow\uparrow}_{1234}(\nu, \nu', \omega) \pm \gamma^{\uparrow\downarrow}_{1234}(\nu, \nu', \omega).$$

Now we can write the first-order, local in site (i) DF-correction to the dual self-energy (Fig.6):

$$\widetilde{\Sigma}^{(1)i}_{12}(\nu) = \sum_{\nu',3,4} \gamma^d_{1234}(\nu, \nu', 0)\widetilde{G}^{ii}_{43}(\nu'). \tag{45}$$
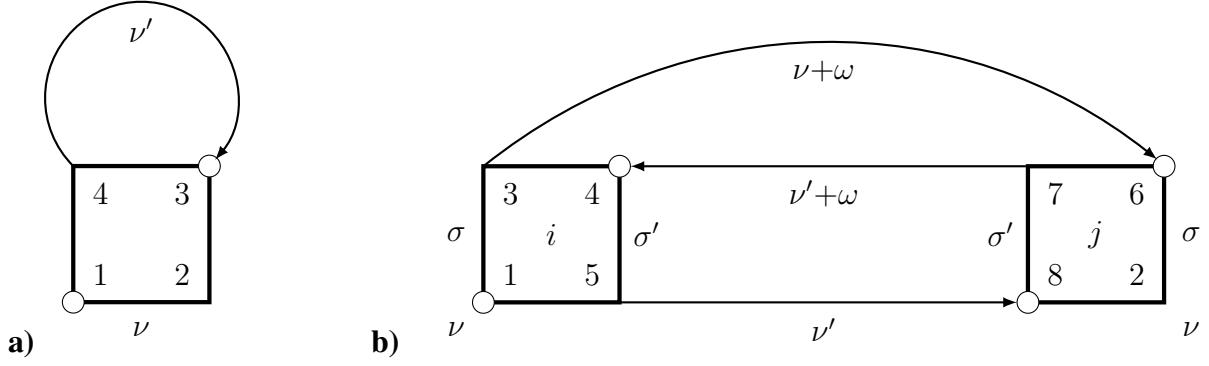
Fig. 6: *Feynman diagram for the 1-st order (a) and the 2-nd order (b) of dual fermion perturbation for the self-energy $\widetilde{\Sigma}$: a line represents the non-local $\widetilde{G}_{43}$ and a box is the local γ_{1234}.*

We now can use the freedom to chose the hybridization function Δ_ν in order to eliminate the main first-oder dual fermion correction Eq. (45) and all other local DF-diagram. Since the vertex function γ_{1234} is purely local, it is enough to ensure that the local part of dual Green function vanishes $\widetilde{G}_{loc} = 0$. This condition is exactly equivalent to the DMFT self-consistent condition for the hybridization function Δ_ν (Fig.7)

$$\sum_{\mathbf{k}} \left(g_\nu^{-1} + \Delta_\nu - t_{\mathbf{k}}\right)^{-1} = g_\nu . \tag{46}$$

The effective impurity model Eq. (30), which is fully determined by the local hybridization function Δ_ν on fermionic Matsubara frequencies $i\nu_n$ is solved using the numerically exact ct-QMC scheme [3] and the exact local Green function g_ν is obtained. The self-consistent DMFT condition for the hybridization function equates the local part of the lattice Green function with that of the impurity, which shows that DMFT minimizes, in a local sense, the distance $|t_{\mathbf{k}} - \Delta_\nu|$. It worthwhile to point here that the "free" or non-interacting dual fermions are equivalent to the full solution of the DMFT problem, This is why dual fermions are only "weakly interacting" so that this perturbation scheme can be very efficient, provided a good reference system.
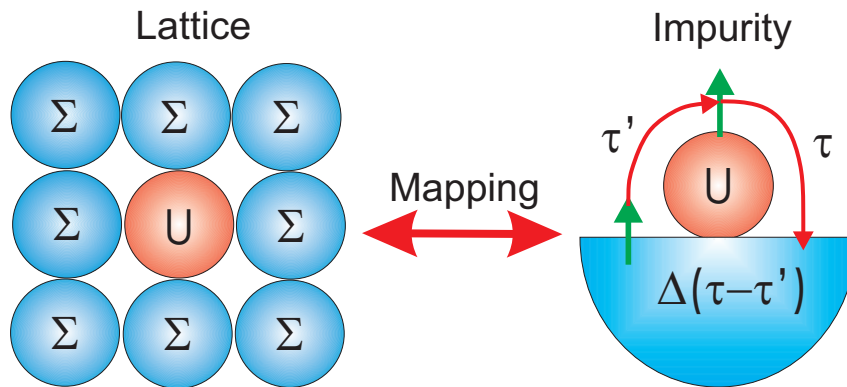
Fig. 7: *Schematic representation of the DMFT reference system for correlated lattice models.*

The second-order Feynman diagram for DF-perturbation (Fig.6) in real space ($\mathbf{R_{ij}}$) has density- and spin-channel contributions with corresponding constants ($c_d = -1/4$ and $c_m = -3/4$)

$$\widetilde{\Sigma}_{12}^{(2)ij}(\nu) = \sum_{\nu'\omega} \sum_{3-8} \sum_{\alpha=d,m} c_\alpha \gamma_{1345}^{\alpha,i}(\nu,\nu',\omega)\, \widetilde{G}_{36}^{ij}(\nu+\omega)\, \widetilde{G}_{74}^{ji}(\nu'+\omega)\, \widetilde{G}_{58}^{ij}(\nu')\, \gamma_{8762}^{\alpha,j}(\nu',\nu,\omega)\,.$$

Using the exact relation between dual Green function and real Green function [5,22], we can express the total lattice self-energy as the sum of the reference contribution Σ^0 (e.g. the impurity) and corrections Σ' which related with the dual self-energy $\widetilde{\Sigma}$

$$\begin{aligned}
\Sigma_{\mathbf{k}\nu} &= \Sigma_\nu^0 + \Sigma'_{\mathbf{k}\nu} \\
\Sigma'_{\mathbf{k}\nu} &= g_\nu^{-1} - \left(g_\nu + \widetilde{\Sigma}_{\mathbf{k}\nu}\right)^{-1}.
\end{aligned}$$

We pointe out that this expression is related with the exact transformation between dual- and real-spaces [4] which has general consequences for similar combinations including strong-coupling techniques, like DMFT or DF.

What is much more important for numerical calculations is that, starting from the full DF-Green function: $\widetilde{G}_{\mathbf{k}\nu}^{-1} = \widetilde{G}_{0\mathbf{k}\nu}^{-1} - \widetilde{\Sigma}_{\mathbf{k}\nu}$ and the exact relation between dual and real Green function [5,22], we can find an expression for the lattice Green function including only the reference impurity Green function and the dual self-energy [4]

$$G_{\mathbf{k}\nu} = \left(\left(g_\nu + \widetilde{\Sigma}_{\mathbf{k}\nu}\right)^{-1} - \widetilde{\Delta}_{\mathbf{k}\nu}\right)^{-1} \tag{47}$$

This formula is perfectly suitable for ct-QMC calculations for realistic multi-orbital correlated matter, where from noisy Monte-Carlo data one only needs local one- and two-particle Green functions, without knowledge of a local self-energy and an "amputated" vertex function.

In principle, one can go beyond second order perturbation theory and include dual ladder diagrams [22], dual parquet diagrams [23] or even try to sum up all dual diagrams with two-particle vertex γ_{1234} stochastically, using a diagrammatic Monte Carlo in dual space [24, 25]. We also can make the diagrammatic series self-consistent, using the "bold" line and update the bare dual Green function with the dual self-energy. Finally, one can also "upgrade" the reference system, which is not anymore "best" for dual fermion theory, but only for the DMFT approach with non-interacting dual fermions.

As numerical example of the DF-perturbation, we discuss a simple, but non-trivial case of a half-field Hubbard model in the strong-coupling case $U = W = 8t$ ($t = 1$, U is equal to the bandwidth $W = 8t$) and the temperature $\beta = 5$. We use a free periodic plaquette as a reference system and perform second-order DF-perturbation. We use exact-diagonalization solver and the Padé analytical continuation to the real axes [7]. Results of the second-order DF superperturbation are shown on Fig. 8 together with numerical exact lattice Monte Carlo results and exact diagonalization of a 4×4 periodic cluster. The DOS for the second order dual fermion plaquette perturbations are in a good agreements with the two numerically exact schemes, ED and DQMC. The DF theory reproduces the so-called four-peak structure of the half-filled Hubbard model, which is a standard feature of lattice QMC calculations [26].
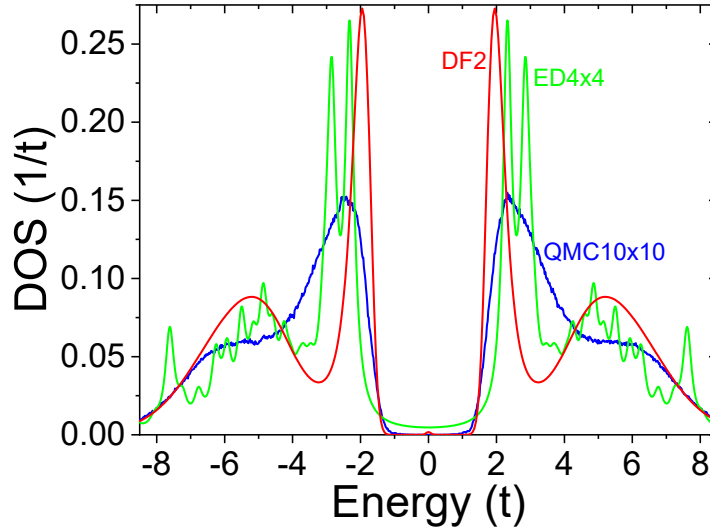
Fig. 8: *Density of states for the half-filled Hubbard model with $U = W = 8t$ in the second-order DF approximation (DF2) in comparison to ED results for a $4{\times}4$ cluster and lattice QMC simulation of a $10{\times}10$ system.*

6 LDA+DMFT scheme for real materials

In order to investigate real correlated systems with the local DMFT scheme we need to have an efficient scheme of partitioning the space and orbital degrees of freedom. For example in the high-temperature superconducting oxide $YBa_2Cu_3O_7$ the strongly correlated electrons are the Cu $3d$'s, and moreover there is only one per non-equivalent copper $d_{x^2-y^2}$ band which crosses the Fermi level with strong many-body fluctuations. Just a few percent of the total number of electronic states need to be included in the DMFT calculations. Therefore the simplest realistic correlated scheme would be a DFT+DMFT approach [27, 28] with partitioning of the orbital space to the normal band electrons $|K\rangle$ described by the DFT Bloch basis and the correlated local orbitals $|L\rangle$ described by some optimal Wannier basis (see Fig. 9 for an illustration).

The treatment of correlated electron systems requires the calculation of Green functions and hybridization functions in terms of local orbitals. This is readily achieved when using a basis set, which is localized in real space, such as linear (or N-th order) muffin-tin orbitals (NMTO) [29], plane-waves [30], Gaussian basis sets [31], or a projector scheme [32]. However, many implementations of density functional theory use a delocalized plane-wave basis set. This has the advantage, that the basis set is simple, universal, and its convergence is controlled in principle by a single parameter, the energy cutoff. The projector augmented wave method (PAW) [33], being a representative of a plane-wave based methods, can be used as a simple example of the general projection scheme from the Bloch to the local basis: $\langle K|L\rangle$ (Fig. 9).

Following the general projection scheme of Ref. [32, 31], the desired quantity for an implementation of the DFT+DMFT method is a projection $\mathcal{P}^{\mathcal{C}} = \sum_L |L\rangle \langle L|$ of the full DFT Kohn-Sham Green function $G_{\mathrm{KS}}(\omega)$ on a set of localized orbitals $\{|L\rangle\}$

$$G^{\mathcal{C}}(\omega) = \mathcal{P}^{\mathcal{C}} G^{\mathrm{KS}}(\omega) P^{\mathcal{C}}. \tag{48}$$
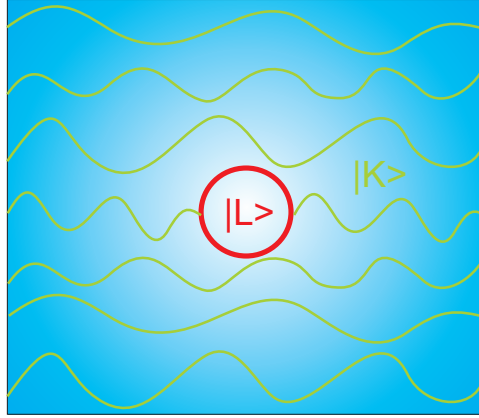
Fig. 9: *Schematic representation of the projection scheme from the Bloch basis to a local Wannier correlated subset.*

The subspace $\mathcal{C} = \mathrm{span}(\{\,|L\rangle\,\})$ is usually called the correlated subspace. It is the subspace of orbitals in which many-body fluctuations play a major role and where the DMFT corrections to the DFT will be considered. In plane-wave based calculations, $G^{\mathrm{KS}}(\omega)$ in Matsubara space is available in terms of an almost complete set of Bloch states $|K\rangle$ that are eigenstates of the Kohn-Sham Hamiltonian $H_{\mathrm{KS}}|K\rangle = \varepsilon_K |K\rangle$

$$G_{\mathrm{KS}}(\omega) = \sum_K \frac{|K\rangle\langle K|}{i\omega + \mu - \varepsilon_K}. \tag{49}$$

Inserting equation (49) into (48) shows that one needs to evaluate projections of type $\langle L|K\rangle$ in order to access the matrix elements $G^{\mathcal{C}}_{LL'}(\omega)$ of the local Green function. In most cases the correlated orbitals are d or f orbitals, which, to a good approximation, are localized inside the PAW augmentation spheres. For $|L\rangle$ within these spheres and given the PAW decomposition of a Bloch state $|K\rangle$ [33] one obtains

$$\langle L|K\rangle = \sum_i \langle L|\varphi_i\rangle\langle \tilde{p}_i|\tilde{K}\rangle. \tag{50}$$

The index i of the augmentation functions $|\varphi_i\rangle$ includes site s, angular momentum l and m, as well as an index ν labeling the radial function: $i = (s, l, m, \nu)$, while the $|\tilde{p}_i\rangle$ are projectors of the PAW scheme.

In the described projections scheme the $|L\rangle\langle L|$ matrices are not properly normalized for two reasons: (1) the Bloch basis is incomplete since only a limited number of Bloch bands is included and (2) the PAW augmentation functions are in general not orthonormal. The simplest way is to orthonormalized the projection matrices by the following Wannier type construction: By definition, the localized states $|L\rangle$ are labeled by site and angular momentum indices: $L = (s, l, m)$. We split the site index $s = \mathbf{R} + \mathbf{T}$ such that $\mathbf{R}$ labels the position within the unit cell and $\mathbf{T}$ is the Bravais lattice vector of the unit cell in which s is located. This allows us to construct the Bloch transform of the localized states,

$$|L_{\mathbf{k}}\rangle = \sum_T e^{i\mathbf{k}\mathbf{T}} |L_{\mathbf{T}}\rangle, \tag{51}$$

where $\mathbf{k}$ is from the first Brillouin zone and $|L_{\mathbf{T}}\rangle \equiv |L\rangle = |s, l, m\rangle$. The sum in equation (51) runs over the Bravais lattice. Labeling the Bloch states $|K\rangle = |\mathbf{k}, n\rangle$ by their crystal momentum, $\mathbf{k}$, and band index, n, we normalize our projection matrices $\mathcal{P}^{\mathcal{C}}_{Ln}(\mathbf{k}) = \langle L_{\mathbf{k}}|\mathbf{k}, n\rangle$ using the overlap operator

$$O_{LL'}(\mathbf{k}) = \sum_n \mathcal{P}^{\mathcal{C}}_{Ln}(\mathbf{k})\, \mathcal{P}^{*\mathcal{C}}_{L'n}(\mathbf{k}) \tag{52}$$

in

$$\bar{\mathcal{P}}^{\mathcal{C}}_{Ln}(\mathbf{k}) = \sum_{L'} O^{-1/2}_{LL'}(\mathbf{k}) P^{\mathcal{C}}_{L'n}(\mathbf{k}). \tag{53}$$

These orthonormalized projection matrices are calculated once at the beginning of any calculation and can then be used to obtain the local Green function of the correlated orbitals from the full Bloch Green function $G^B_{nn'}$

$$G^{\mathcal{C}}_{LL'}(\omega) = \sum_{\mathbf{k}, nn'} \bar{\mathcal{P}}^{\mathcal{C}}_{Ln}(\mathbf{k})\, G^B_{nn'}(\mathbf{k}, \omega)\, \bar{\mathcal{P}}^{*\mathcal{C}}_{L'n'}(\mathbf{k}).$$

Similarly the hybridization function, $\Delta(\omega)$, is available. It is related to the local Green function by

$$G^{-1}(\omega) = i\omega - \epsilon_d - \Delta(\omega), \tag{54}$$

where ϵ_d is the static crystal field. Equation (54) is a matrix equation with G, Δ, and ϵ_d being $(\dim \mathcal{C}) \times (\dim \mathcal{C})$ matrices, in general. To separate the hybridization from the static DFT crystal field, we numerically evaluate the limit $\omega \to \infty$, where $\omega - G^{-1}(\omega) \to \epsilon_d$.

In a DFT+DMFT calculation the projection matrices $\bar{\mathcal{P}}^{\mathcal{C}}_{Ln}(k)$ are used for up- and downfolding quantities like the Green function and the self energy in the course of the iterative DMFT procedure in exactly the same way as shown for the local Green function above. For example, the self energy obtained by an impurity solver for the effective impurity model $\Sigma^{\mathcal{C}}_{LL'}(\omega)$ can be upfolded to the Bloch basis as

$$\Sigma^B_{nn'}(\mathbf{k}, \omega) = \sum_{LL'} \overline{P}^{*\mathcal{C}}_{Ln}(\mathbf{k})\, \Sigma^{\mathcal{C}}_{LL'}(\omega)\, \overline{P}^{\mathcal{C}}_{L'n'}(\mathbf{k}).$$

Since the self energy in DMFT is a purely local quantity, the index $\mathbf{k}$ on $\Sigma^B_{nn'}(\mathbf{k}, \omega)$ reflects the momentum dependence brought about by the projection matrices. The presented projection scheme allows for the inclusion of both correlated and uncorrelated states in the procedure. Therefore, information about the interplay of correlated orbitals with their uncorrelated ligands can be obtained.

Figure 10 shows that the DFT+DMFT calculation commences with the solution of the Kohn-Sham equations of DFT. In a second step the projection onto the correlated subset is computed. The Kohn-Sham Green function is then computed and used as an initial guess for the mean-field $\mathcal{G}$ of the DMFT cycle, which consists of the usual steps detailed before. In usual applications, the DFT+DMFT loop will stop after DMFT self-consistency is obtained. It has, however, recently become possible to continue the cycle supplying the DFT code with an altered charge density that includes correlation effects. In such a unified approach, changes in the charge
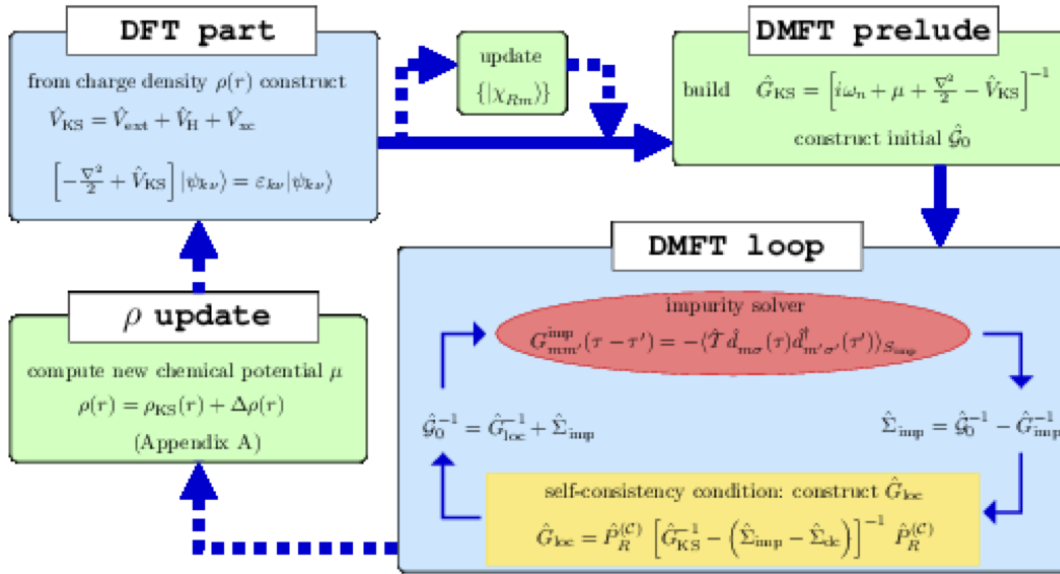
Fig. 10: *Illustration of the DFT+DMFT procedure. As a first step, the Kohn-Sham (KS) equations, determining the Kohn-Sham potential and thus the Hamiltonian, are solved. Secondly, the KS Green function and from it the starting value for the bath Green function $\mathcal{G}$ is constructed and passed on to the DMFT loop, which consists of the usual steps described before. A potential self-consistency over the charge density is also indicated [31].*

density induced by correlations can be studied [31, 34]. It allows furthermore for the accurate calculation of total energies, that allow the determination of crystal structures and other coupled electronic and structural effects. We will briefly sketch here the most important elements of the charge self-consistent implementation of DFT+DMFT following Ref. [31].

As example, we show a realistic DFT+DMFT calculation of ferromagnetic spectral function for Fe and Ni at $T = 380$ K in Fig. 11 [35], in comparison with experimental ARPES spectra for Fe [36] and Ni [37]. For the DFT part the VASP code with the Wannier projector scheme was used [32] and for DMFT impurity solver numerical exact ct-QMC was used [3]. One can see a good agreement for renormalization of the band dispersion near the Fermi level for Ni, but some discrepancy for Fe (specially in P-H-Γ direction). This indicates that one needs to include non-local self-energy effects in Fe beyond the DMFT, such as in the multi-orbital DF-scheme.

We do not discuss the problem of "double counting" in the LDA+DMFT formalism: since any DFT approximation includes average of Coulomb interactions, one should subtract these from the DMFT part. In practice it is very hard to fulfill, since we do not know this part analytically and normally we chose the double counting correction from the LDA+U scheme as a static version of LDA+DMFT [27, 28]. The main problem is related with the fact that both DFT and DMFT are non-perturbative, strong-coupling theories which need to be "aligned" on some reference level, like the chemical potential. One can try to search for this "reference level" as a free parameter [38] and compare the resulting spectral function of the LDA+DMFT with experimental photoemission spectrum. Another possibility is to start from an analytical
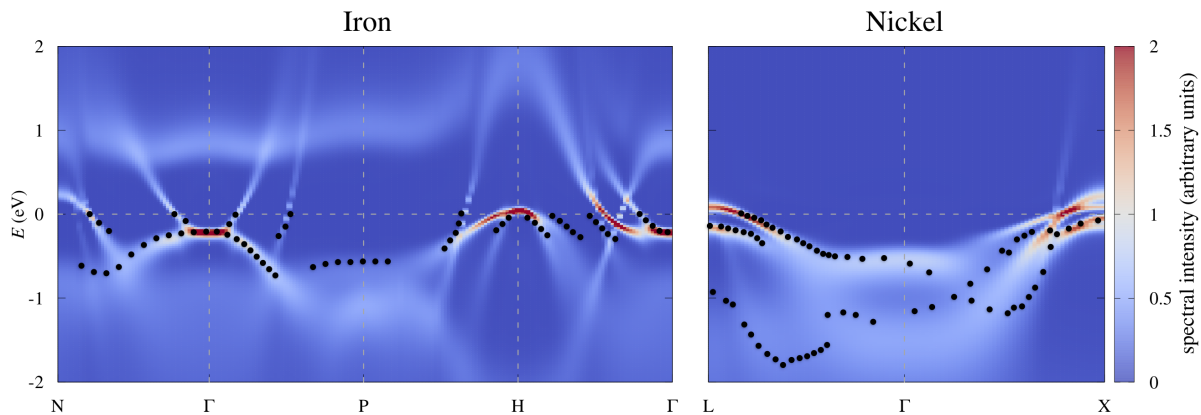
Fig. 11: *Momentum resolved spectral function for ferromagnetic Fe and Ni obtained by LDA+DMFT for T=380 K in comparison with photoemission data (dots) [35].*

many-body scheme like GW and design a GW+DMFT approach where one knows the local correlated part analytically [39]. If we would like to incorporate non-local self-energy effects beyond the DMFT, then such a simple "double counting" schemes may not work any more and more elaborat dual fermion/boson schemes should be considered. There is also the complicated problem of estimating the effective local Coulomb matrix elements in the Wannier basis used in LDA+DMFT. One of the most accurate schemes is related with a constrained RPA scheme [40]. We can mention a recent discussion of the cRPA scheme for strongly correlated insulators [41]. One of the recent accurate charge self-consistent LDA+DMFT software packages is based on combination of Quantum ESPRESSO (DFT), Wannier90 (Projectors), and TRIQS (ct-QMC) [42].

7   Conclusions

We have discussed the path integral expansion for general reference systems which help to understand the DFT scheme and the correlated DMFT approach for interacting lattice fermions and their combination for realistic systems. The transformation to dual variables allowed to go beyond the local DMFT approximation for lattice self-energy. The accurate multi-orbital DFT+DMFT scheme for strongly correlated transition metals and rare earth systems can described the spin, orbital and charge fluctuations in the d- or f-shell which play the crucial role in photoemission spectra, as well as magnetic and optical excitations. The numerically exact ct-QMC solution of the quantum impurity problem gives an effective local exchange-correlation functional for given correlated materials in a specific external hybridization. The accurate combination of the DMFT scheme with first-principle DFT approaches gives a very useful tool for the investigation of correlated electronic materials.

A   Path integrals for fermions

We first introduce a formalism of the path integral over fermionic fields [15]. Let us consider a simple case of a single quantum state $|i\rangle$ occupied by fermionic particles [16]. Due to the Pauli principle the many-body Hilbert space is spanned by only two orthonormal states $|0\rangle$ and $|1\rangle$. In the second quantization scheme for fermions with annihilation $\hat{c}_i$ and creation $\hat{c}_i^\dagger$ operators with anticommutation relations $\{\hat{c}_i, \hat{c}_j^\dagger\} = \delta_{ij}$ we have the following simple rules

$$\hat{c}_i\,|1\rangle = |0\rangle \quad \hat{c}_i\,|0\rangle = 0 \quad \text{and} \quad \hat{c}_i^\dagger\,|0\rangle = |1\rangle \quad \hat{c}_i^\dagger\,|1\rangle = 0\,. \tag{55}$$

Moreover, the density operator and the Pauli principle have the form

$$\hat{c}_i^\dagger \hat{c}_i\,|n\rangle = n_i\,|n\rangle \quad \text{and} \quad \hat{c}_i^2 = (\hat{c}_i^\dagger)^2 = 0\,.$$

The central object here are the so-called fermionic coherent states $|c\rangle$, which are eigenstates of annihilation operator $\hat{c}_i$ with eigenvalue c_i

$$\hat{c}_i\,|c\rangle = c_i\,|c\rangle\,. \tag{56}$$

It is worthwhile to note that such a left-eigenbasis has only annihilation operators, due to the fact that they are bounded from the below and one can rewrite one of equations from Eq. (55) in the following "eigenvalue" form

$$\hat{c}_i\,|0\rangle = 0\,|0\rangle\,.$$

Due to the anti-commutation relations for the fermionic operators the eigenvalues of coherent states c_i are so-called Grassmann numbers with the multiplication rules [17]

$$c_i c_j = -c_j c_i \quad \text{and} \quad c_i^2 = 0\,. \tag{57}$$

It is convenient to assume that the Grassmann numbers also anti-commute with the fermionic operators

$$\{c, \hat{c}\} = \{c, \hat{c}^\dagger\} = 0\,.$$

An arbitrary function of one Grassmann variable can be represented by only the first two Taylor coefficients

$$f(c) = f_0 + f_1 c\,. \tag{58}$$

One can prove the following general many-body representation of coherent states

$$|c\rangle = e^{-\sum_i c_i \hat{c}_i^\dagger}\,|0\rangle\,. \tag{59}$$

Let us show this for the simple case of one fermionic state

$$\hat{c}\,|c\rangle = \hat{c}\big(1 - c\hat{c}^\dagger\big)\,|0\rangle = \hat{c}\big(\,|0\rangle - c\,|1\rangle\,\big) = -\hat{c}c\,|1\rangle = c\,|0\rangle = c\,|c\rangle\,. \tag{60}$$

One can also define a "left" coherent state $\langle c|$ as the left-eigenstates of creation operators $\hat{c}_i^\dagger$

$$\langle c|\,\hat{c}_i^\dagger = \langle c|\,c_i^*\,.$$

Note that the new eigenvalue c_i^* is just another Grassmann number, not the complex conjugate of c_i. The left coherent state can be obtained similar to Eq. (59)

$$\langle c| = \langle 0| \, e^{-\sum_i \hat{c}_i c_i^*}.$$

A general function of two Grassmann variables can, analogously to Eq. (58), be represented by only four Taylor coefficients

$$f(c^*, c) = f_{00} + f_{10}c^* + f_{01}c + f_{11}c^*c. \tag{61}$$

Using this expansion we can define a derivative of Grassmann variables in the natural way

$$\frac{\partial c_i}{\partial c_j} = \delta_{ij}.$$

One needs to be careful with the "right order" of such a derivative and remember the anti-commutation rules, i.e.,

$$\frac{\partial}{\partial c_2} c_1 c_2 = -c_1.$$

For the case of the general two-variable function in Eq. (61) we have

$$\frac{\partial}{\partial c^*} \frac{\partial}{\partial c} f(c^*, c) = \frac{\partial}{\partial c^*} \left(f_{01} - f_{11}c^* \right) = -f_{11} = -\frac{\partial}{\partial c} \frac{\partial}{\partial c^*} f(c^*, c).$$

One also needs a formal definition of the integration over Grassmann variables, and the natural way consists of the following rules [17]

$$\int 1 \, dc = 0 \quad \text{and} \quad \int c \, dc = 1 \,,$$

which just shows that the integration over a Grassmann variable is equivalent to differentiation

$$\int \cdots dc \rightarrow \frac{\partial}{\partial c} \cdots$$

The coherent states are not orthonormal and the overlap of any two such states is equal to

$$\langle c|c \rangle = e^{\sum_i c_i^* c_i}$$

which is easy to see for the case of one particle

$$\langle c|c \rangle = \left(\langle 0| - \langle 1| \, c^* \right) \left(|0\rangle - c \, |1\rangle \right) = 1 + c^*c = e^{c^*c}.$$

An important property of coherent states is the resolution of unity

$$\int dc^* \int dc \, e^{-\sum_i c_i^* c_i} \, |c\rangle\langle c| = \hat{1} = \iint dc^* dc \, \frac{|c\rangle\langle c|}{\langle c|c\rangle}.$$

For simplicity we demonstrate this relation only for one fermion state

$$\iint dc^* dc \, e^{-c^*c} \, |c\rangle\langle c| = \iint dc^* dc \, \left(1 - c^*c \right) \left(|0\rangle - c \, |1\rangle \right) \left(\langle 0| - \langle 1| \, c^* \right)$$

$$= -\iint dc^* dc \, c^*c \, \left(|0\rangle\langle 0| + |1\rangle\langle 1| \right) = \sum_n |n\rangle\langle n| = \hat{1} \,.$$

Matrix elements of normally ordered operators are very easy to calculate in the coherent basis by operating with $\hat{c}^\dagger$ on the states to the right and $\hat{c}$ to the left:

$$\langle c^* | \, \hat{H}(\hat{c}^\dagger, \hat{c}) \, | c \rangle = H(c^*, c) \, \langle c^* | c \rangle \, . = H(c^*, c) \, e^{\sum_i c_i^* c_i} \tag{62}$$

Within the manifold of coherent states we can map the fermionic operators to the Grassmann variables $(\hat{c}_i^\dagger, \hat{c}_i) \rightarrow (c_i^*, c_i)$.

Finally, we prove the so-called "trace-formula" for arbitrary fermionic operators in normal order (in one-fermion notation)

$$\mathrm{Tr}\,\hat{O} = \sum_{n=0,1} \langle n | \, \hat{O} \, | n \rangle = \sum_{n=0,1} \iint dc^* dc \, e^{-c^* c} \, \langle n | \, c \rangle \langle c | \, \hat{O} \, | n \rangle =$$

$$= \iint dc^* dc \, e^{-c^* c} \sum_{n=0,1} \langle -c | \, \hat{O} \, | n \rangle \langle n | \, c \rangle = \iint dc^* dc \, e^{-c^* c} \, \langle -c | \, \hat{O} \, | c \rangle \, .$$

The fermionic "minus" sign in the left coherent states come from the commutation of the (c^*) and (c) coherent state in such a transformation: $\langle n|c \rangle \, \langle c|n \rangle = \langle -c|n \rangle \, \langle n|c \rangle$. One has to use the standard Grassmann rules: $c_i^* c_j = -c_j c_i^*$ and $|-c \rangle = |0 \rangle + c \, |1 \rangle$.

We are ready now to write the partition function for the grand-canonical quantum ensemble with $H = \hat{H} - \mu \hat{N}$ and inverse temperature β. One has to use the N-slices Trotter decomposition for the partition function in $[0, \beta)$ with imaginary time $\tau_n = n\Delta\tau = n\beta/N$ ($n = 1, ..., N$), and insert N times the resolution of unity as follows

$$Z = \mathrm{Tr}\, e^{-\beta H} = \iint dc^* dc \, e^{-c^* c} \langle -c | e^{-\beta H} | c \rangle$$

$$= \int \Pi_{n=1}^N dc_n^* dc_n \, e^{-\sum_n c_n^* c_n} \langle c_N | \, e^{-\Delta\tau H} \, | c_{N-1} \rangle \langle c_{N-1} | \, e^{-\Delta\tau H} \, | c_{N-2} \rangle \, ... \, \langle c_1 | \, e^{-\Delta\tau H} \, | c_0 \rangle$$

$$= \int \Pi_{n=1}^N dc_n^* dc_n \, e^{-\Delta\tau \sum_{n=1}^N \left(c_n^* (c_n - c_{n-1})/\Delta\tau + H(c_n^*, c_{n-1}) \right)}$$

In the continuum limit $(N \rightarrow \infty)$

$$\Delta\tau \sum_{n=1}^N \cdots \rightarrow \int_0^\beta d\tau \cdots, \qquad \frac{c_n - c_{n-1}}{\Delta\tau} \rightarrow \partial_\tau \quad \text{and} \quad \Pi_{n=0}^{N-1} dc_n^* dc_n \rightarrow \mathcal{D}\,[c^*, c]$$

with antiperiodic boundary conditions for fermionic Grassmann variables in imaginary time $c(\tau)$ and $c^*(\tau)$

$$c(\beta) = -c(0), \qquad c^*(\beta) = -c^*(0)$$

we end up in the standard path-integral formulation of the partition function

$$Z = \int \mathcal{D}\,[c^*, c] \, e^{-\int_0^\beta d\tau \left(c^*(\tau)\partial_\tau c(\tau) + H(c^*(\tau), c(\tau)) \right)} \, . \tag{63}$$

It is useful to mention the general form of the Gaussian path-integral for a non-interacting "quadratic" fermionic action, which is equivalent to the Hubbard-Stratonovich transformation

used in the dual-fermion derivation Eq. (32). For an arbitrary matrix M_{ij} and Grassmann vectors J_i^* and J_i one can calculate analytically the following integral

$$Z_0\left[J^*, J\right] = \int \mathcal{D}\left[c^*c\right] e^{-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j - \sum_{i=1}^{N}\left(c_i^* J_i + J_i^* c_i\right)} = \det M \, e^{\sum_{i,j=1}^{N} J_i^* (M^{-1})_{ij} J_j} \, . \quad (64)$$

To prove this relation one needs to first change variables in order to eliminate J_i^* and J_i and expand the exponential function (only the N-th oder is non-zero)

$$e^{-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j} = \frac{1}{N!}\left(-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j\right)^N \, .$$

Finally, different permutations of c_i^* and c_j and integration over Grassmann variables will give $\det M$. As a small exercise we will check such integrals for the first two many-particle dimensions. For $N=1$ it is trivial

$$\int \mathcal{D}\left[c^*c\right] e^{-c_1^* M_{11} c_1} = \int \mathcal{D}\left[c^*c\right]\left(-c_1^* M_{11} c_1\right) = M_{11} = \det M$$

and for $N=2$ we have

$$\int \mathcal{D}\left[c^*c\right] e^{-c_1^* M_{11} c_1 - c_1^* M_{12} c_1 - c_2^* M_{21} c_1 - c_2^* M_{22} c_2} =$$

$$= \frac{1}{2!}\int \mathcal{D}\left[c^*c\right]\left(-c_1^* M_{11} c_1 - c_1^* M_{12} c_1 - c_2^* M_{21} c_1 - c_2^* M_{22} c_2\right)^2 = M_{11} M_{22} - M_{12} M_{21} = \det M.$$

For a change of variables in the path integral one uses the following transformation with unit Jacobian: $c \to c + M^{-1} J$ and

$$c^* M c + c^* J + J^* c = \left(c^* + J^* M^{-1}\right) M \left(c + M^{-1} J\right) - J^* M^{-1} J \, .$$

Using the Gaussian path-integral it is very easy to calculate any correlation function for a non-interaction action (Wick-theorem)

$$\left\langle c_i c_j^*\right\rangle_0 = -\frac{1}{Z_0}\frac{\delta^2 Z_0\left[J^*, J\right]}{\delta J_i^* \, \delta J_j}\bigg|_{J=0} = M_{ij}^{-1}$$

$$\left\langle c_i c_j c_k^* c_l^*\right\rangle_0 = \frac{1}{Z_0}\frac{\delta^4 Z_0\left[J^*, J\right]}{\delta J_i^* \delta J_j^* \delta J_l \delta J_k}\bigg|_{J=0} = M_{il}^{-1} M_{jk}^{-1} - M_{ik}^{-1} M_{jl}^{-1}.$$

Corresponding bosonic path-integrals can be formulated in a similar way with complex variables and periodic boundary conditions on imaginary time. The Gaussian path-integral over bosonic fields is equal to inverse of the M-matrix determinant [15].

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