

The DMFT object of the CP-PAW code

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Chapter 1

Purpose and theoretical background

The purpose of the DMFT object is to prepare an interface to the solver for a quantum impurity, in the context of dynamical mean-field theory.

1.1 Grand potential and density-matrix functional as starting point

The **grand potential** has the form[1]

$$\Omega_{\beta,\mu}^{KB}[\hat{h} + \hat{W}] = \min_{|\psi_n\rangle, f_n \in [0,1]} \text{stat}_{\Lambda} \left\{ \sum_n f_n \langle \psi_n | \hat{h} | \psi_n \rangle + \tilde{F}_{\beta}^{\hat{W}} \left[\sum_n |\psi_n\rangle f_n \langle \psi_n| \right] - \mu \sum_n f_n - \sum_{m,n} \Lambda_{m,n} \left(\langle \psi_n | \psi_m \rangle - \delta_{m,n} \right) \right\} \quad (1.1)$$

where the **density-matrix functional** is expressed with the **Luttinger-Ward functional** [2] $\Phi^{LW}[\mathbf{G}, \hat{W}]$ as

$$\begin{aligned} \tilde{F}_{\beta}^{\hat{W}}[\rho] &= \frac{1}{\beta} \text{Tr} \left[\rho \ln(\rho) + (1 - \rho) \ln(1 - \rho) \right] \\ &+ \text{stat}_{\mathbf{h}'} \text{stat}_{\mathbf{G}, \Sigma} \left\{ \Phi_{\beta}^{LW}[\mathbf{G}, \hat{W}] - \frac{1}{\beta} \sum_{\nu} \text{Tr} \left\{ \ln \left[\mathbf{1} - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' + \Sigma(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \right\} \right. \\ &\quad \left. + \left(\mathbf{h}' + \Sigma(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \mathbf{G}(i\omega_{\nu}) - \left[\mathbf{G}(i\omega_{\nu}) - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' - \mathbf{h}_{\rho} \right) \right\} \end{aligned} \quad (1.2)$$

Here, $|\psi_n\rangle$ are one-particle wave functions. They play the role of **natural orbitals**, the eigenstates of the **one-particle density matrix** ρ . f_n are the occupations, the eigenvalues of the one-particle density matrix. The chemical potential is μ and $\beta = 1/(k_B T)$. The full Hamiltonian consists of a non-interacting part $\hat{h} = \frac{\hat{p}^2}{2m_0} + \hat{v}_{ext}$ and an electron-electron interaction \hat{W} .

The Matsubara sum runs over the **Matsubara frequencies** (see appendix A on p. 22)

$$\omega_{\nu} = (2\nu - 1) \frac{\pi}{\hbar\beta} \quad \text{for } \nu \in \mathbb{Z} \quad (1.3)$$

The Hamiltonian \mathbf{h}_ρ is a non-local Hamiltonian directly related to the one-particle density matrix via

$$\mathbf{h}_\rho = \mu \mathbf{1} + k_B T \ln \left[\frac{\mathbf{1} - \rho}{\rho} \right] \quad (1.4)$$

The Hamiltonian \mathbf{h}' , on the other hand, is a Lagrange multiplier.

$\mathbf{G}(i\omega)$ is the Green's function and $\Sigma(i\omega_\nu)$ is the self energy. The Green's function is related to the density matrix by

$$\rho = \frac{1}{\beta} \sum_{\nu} e^{i\beta \hbar \omega_{\nu} 0^+} \mathbf{G}(i\omega_{\nu}) \quad (1.5)$$

This latter equation is a consequence of the stationarity with respect to the Lagrange multiplier \mathbf{h}'

In practice, we calculate the **Helmholtz potential**

$$\begin{aligned} A_{\beta, N}[\hat{h} + \hat{W}] &= \text{stat}_{\mu} \left\{ \Omega_{\beta, \mu} + \mu N_{\mu} \right\} \\ &= \min_{|\psi_n\rangle, f_n \in [0, 1]} \text{stat}_{\mu, \Lambda} \left\{ \sum_n f_n \langle \psi_n | \hat{h} | \psi_n \rangle + Q_{\beta}^{\hat{W}} \left[\sum_n |\psi_n\rangle f_n \langle \psi_n| \right] \right. \\ &\quad \left. + \frac{1}{\beta} \sum_n \left[f_n \ln(f_n) + (1 - f_n) \ln(1 - f_n) \right] \right. \\ &\quad \left. - \mu \left[\sum_n f_n - N \right] - \sum_{m, n} \Lambda_{m, n} \left(\langle \psi_n | \psi_m \rangle - \delta_{m, n} \right) \right\} \end{aligned} \quad (1.6)$$

where Q is the density matrix functional without the entropy contribution. The entropy contribution is taken care of with the Mermin functional[3] to describe DFT calculations with electrons at finite temperature.

$$\begin{aligned} Q_{\beta}^{\hat{W}}[\rho] &= \tilde{F}_{\beta}^{\hat{W}}[\rho] - \frac{1}{\beta} \text{Tr} \left[\rho \ln(\rho) + (\mathbf{1} - \rho) \ln(\mathbf{1} - \rho) \right] \\ &= \text{stat}_{\mathbf{h}'} \text{stat}_{\mathbf{G}, \Sigma} \left\{ \Phi_{\beta}^{LW}[\mathbf{G}, \hat{W}] - \frac{1}{\beta} \sum_{\nu} \text{Tr} \left\{ \ln \left[\mathbf{1} - \left(i\hbar \omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' + \Sigma(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \right\} \right. \\ &\quad \left. + \left(\mathbf{h}' + \Sigma(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \mathbf{G}(i\omega_{\nu}) - \left[\mathbf{G}(i\omega_{\nu}) - \left(i\hbar \omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' - \mathbf{h}_{\rho} \right) \right\} \end{aligned} \quad (1.7)$$

1.2 Projection onto local orbitals

In order to integrate DMFT into the DFT code, we define first a local basis set of orbitals $|\chi_a\rangle$. These orbitals are not orthogonal. The orbitals are spin orbitals, that is, each is a two-component wave function with a spin-up and a spin-down component. If the spin orbitals are eigenstates of \hat{S}_z , one or the other of the components vanishes.

The decomposition of the Kohn-Sham wave functions, which in the the context of rDMFT are the natural orbitals, is obtained via the projector functions $\langle \pi_a |$ as

$$|\psi_n\rangle = \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle + |\delta \psi_n\rangle \quad (1.8)$$

where the projector functions obey the bi-orthogonality condition

$$\langle \pi_a | \chi_b \rangle = \delta_{a,b} , \quad (1.9)$$

and where $|\delta\psi_n\rangle$ is a remainder which is left over if the local orbitals do not form a complete basis set. This remainder has the property

$$\langle \pi_a | \delta\psi_n \rangle = 0 \quad (1.10)$$

The interaction \hat{W}

$$\hat{W} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_c \hat{c}_d \quad (1.11)$$

is expressed by the **U-tensor**

$$U_{a,b,c,d} = \sum_{\sigma,\sigma'} \int d^3r \int d^3r' \frac{e^2 \chi_a^*(\vec{r}, \sigma) \chi_b^*(\vec{r}', \sigma') \chi_c(\vec{r}, \sigma) \chi_d(\vec{r}', \sigma')}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \quad (1.12)$$

The U-tensor can then be approximated to yield \hat{W}_1 and \hat{W}_2 . Typically, these approximations amount to multiplying matrix elements with scale factors and to leaving certain elements out completely.

Non-orthonormal orbitals

The main difference[4] between orthonormal and non-orthonormal basisets is that the commutator relation

$$\left[\hat{c}_a^\dagger, \hat{c}_b \right]_+ = \langle \pi_b | \pi_a \rangle \quad (1.13)$$

Therefore, it is advisable to first orthonormalize the local orbital basiset.

1.3 DFT and many-particle corrections

In order to integrate correlations into a DFT-like framework, we split off the Helmholtz potential of a DFT calculation, so that the Helmholtz potential has the form of a DFT term and a correction.

$$\begin{aligned} A_{\beta,N}[\hat{h} + \hat{W}] = & \min_{|\psi_n\rangle, f_n \in [0,1]} \text{stat}_{\mu,\Lambda} \left\{ \sum_n f_n \langle \psi_n | \frac{\hat{p}^2}{2m_e} | \psi_n \rangle + \int d^3r n(\vec{r}) v_{\text{ext}}(\vec{r}) \right. \\ & + \frac{1}{2} \int d^3r \int d^3r' \frac{e^2 n(\vec{r}) n(\vec{r}')}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} + E_{\text{xc}}[n] + \frac{1}{\beta} \sum_n \left[f_n \ln(f_n) + (1 - f_n) \ln(1 - f_n) \right] \\ & + \underbrace{Q_{\text{X},\beta}^{\hat{W}_1}[\rho] - Q_{\text{DFT},\beta}^{\hat{W}_1}[\rho, n]}_{\text{local hybrid functional}} + \underbrace{Q_{\beta}^{\hat{W}_2}[\rho] - Q_{\text{X},\beta}^{\hat{W}_2}[\rho]}_{Q_{\text{dyn},\beta}^{\hat{W}_2}[\rho]} \\ & \left. - \mu \left[\sum_n f_n - N \right] - \sum_{m,n} \Lambda_{m,n} \left(\langle \psi_n | \psi_m \rangle - \delta_{m,n} \right) \right\} \quad (1.14) \end{aligned}$$

where

$$\begin{aligned}
n(\vec{r}, \sigma, \sigma') &= \sum_n \langle \vec{r}, \sigma | \psi_n \rangle f_n \langle \psi | \vec{r}, \sigma' \rangle \\
n(\vec{r}) &= \sum_{\sigma} n(\vec{r}, \sigma, \sigma) \\
\rho_{a,b} &= \sum_n \langle \pi_a | \psi_n \rangle f_n \langle \psi | \pi_b \rangle
\end{aligned} \tag{1.15}$$

We distinguish two different corrections:

1. a screened Hartree-Fock correction with an interaction \hat{W}_1 . For this purpose we restrict the U-tensor to onsite contributions and certain bond-terms. In the spirit of the hybrid functionals or the GW method, we scale the U-tensor. The neglect of U-tensor elements that do not reside on an atom pair, is motivated by the fact that screening becomes stronger with increasing distance. Part of this approximation is to neglect the interactions not captured by the local orbital basis. This term is evaluates in the `paw_1mto`-object.
2. a dynamic correction using an interaction \hat{W}_2 . In the spirit of the local approximation the Interaction of this term is limited to on-site terms only. Only the dynamic correlations requires expensive many-particle calculations.

By formulating these terms in correction, we ensure that an approximation of the U-tensor only affects a “small” correction and does not impact the main result.

1.3.1 DFT double-counting correction

The basic ideas behind the double counting correction has been described in a previous paper[4].

1.3.2 Screened Hartree Fock correction

The Hartree-Fock term $Q_{X,\beta}^{\hat{W}}$ is equal to $Q_{\beta}^{\hat{W}}$ when only the first-order term of the Luttinger-Ward functional in the interaction is considered. It is obtained as

$$Q_{X,\beta}^{\hat{W}}[\rho] = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} [\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b}] \tag{1.16}$$

The interpretation of this term is subtle because it is formulated in non-orthonormal orbitals.

- We consider the expansion of Kohn-Sham orbitals in local orbitals

$$\begin{aligned}
Q_{X,\beta}^{\hat{W}}[\rho] &= \frac{1}{2} \sum_{m,n} f_m f_n \int d^3r \int d^3r' \frac{e^2 \left(\phi_m^*(\vec{r}) \phi_n^*(\vec{r}') \phi_n(\vec{r}') \phi_m(\vec{r}) - \phi_m^*(\vec{r}) \phi_n^*(\vec{r}') \phi_m(\vec{r}') \phi_n(\vec{r}) \right)}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \\
&= \frac{1}{2} \sum_{a,b,c,d} \underbrace{\left(\sum_m \langle \pi_a | \phi_m \rangle f_m \langle \phi_m | \pi_b \rangle \right)}_{\rho_{a,b}} \underbrace{\left(\sum_n \langle \pi_c | \phi_n \rangle f_n \langle \phi_n | \pi_d \rangle \right)}_{\rho_{c,d}} \\
&\quad \times \int d^3r \int d^3r' \frac{e^2 \left(\chi_b^*(\vec{r}) \chi_d^*(\vec{r}') \chi_c(\vec{r}') \chi_a(\vec{r}) - \chi_b^*(\vec{r}) \chi_d^*(\vec{r}') \chi_a(\vec{r}') \chi_c(\vec{r}) \right)}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \quad (1.17)
\end{aligned}$$

$$= \frac{1}{2} \sum_{a,b,c,d} \rho_{a,b} \rho_{c,d} \left(U_{b,d,a,c} - U_{b,d,c,a} \right) \quad (1.18)$$

$$= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left(\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \right) \quad (1.19)$$

- Consider expectation value of a Slater determinant expressed in terms of non-orthonormal local orbitals. The Slater determinant has the form

$$\Psi(\vec{x}_1, \dots, \vec{x}_N) = C \det |\mathbf{M}| = C \sum_{i_1, \dots, i_N=1}^N \epsilon_{i_1, \dots, i_N} \chi_{i_1}(\vec{x}_1) \cdots \chi_{i_N}(\vec{x}_N) \quad (1.20)$$

with $M_{ij} = \chi_i(\vec{x}_j)$ and the normalization constant C . $\epsilon_{i_1, \dots, i_N}$ is the fully antisymmetric tensor defined by $\epsilon_{1,2,\dots,N} = 1$, by the fact that it changes sign under permutation of two indices, and that it vanishes whenever two indices are identical.

This argument needs to be completed. Probably we obtain the same result as in the first case. This needs to be shown by performing a transformation onto orthogonal one-particle states, that span the same Hilbert space.

1.3.3 Dynamic correlation correction

The dynamical term $Q_{\text{dyn},\beta}^{\hat{W}}$ is simply the difference of the complete term minus the Hartree-Fock contribution.

DYNAMIC CORRELATION CORRECTION

$$\begin{aligned}
Q_{\text{dyn},\beta}^{\hat{W}}[\rho] &= \tilde{F}_{\beta}^{\hat{W}}[\rho] - \frac{1}{\beta} \text{Tr} \left[\rho \ln(\rho) + (\mathbf{1} - \rho) \ln(\mathbf{1} - \rho) \right] - \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left[\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \right] \\
&= \text{stat}_{\mathbf{h}'} \text{stat}_{\mathbf{G}, \mathbf{\Sigma}} \left\{ \Phi_{\beta}^{LW}[\mathbf{G}, \hat{W}] - \frac{1}{\beta} \sum_{\nu} \text{Tr} \left\{ \ln \left[\mathbf{1} - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' + \mathbf{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \right. \right. \\
&\quad \left. \left. + \left(\mathbf{h}' + \mathbf{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \mathbf{G}(i\omega_{\nu}) - \left[\mathbf{G}(i\omega_{\nu}) - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' - \mathbf{h}_{\rho} \right) \right\} \right\} \\
&\quad - \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left[\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \right] \quad (1.21)
\end{aligned}$$

If the Luttinger-Ward functional is expressed by Feynman diagrams, we can simply avoid the Hartree and the exchange term instead of subtracting the exchange contribution externally. The reason is that we may add any function in a constrained optimization that is constant when the constraint is obeyed. The advantage of this procedure is that the remaining functional depends much weaker on the Green's function.

The purpose of the DMFT object is to add

$$Q_{\text{dyn},\beta}^{\hat{W}_2}[\rho] \quad (1.22)$$

in the local approximation, that is for a Luttinger-Ward functional, that is a sum over local terms. This approximation is consistent with dynamical mean-field theory. In the long run, the DMFT object will only contribute the dynamical terms, while the Hartree-Fock term is taken care of in the LMTO object. This will allow to include the non-local Hartree-Fock terms, while the DMFT object is limited to site local terms for the Luttinger-Ward functional.

1.4 Solver for dynamic correlations

1.4.1 Evaluation of the functional

We evaluate the functional $Q_{\text{dyn},\beta}^{\hat{W}_2}[\rho]$ following the recipe provided in section III of the BPP paper[1].

1. Construct the hamiltonian \hat{h}_ρ , whose Green's function obeys the density-matrix constraint.

$$\hat{h}_\rho = \mu \mathbf{1} + k_B T \ln \left[\frac{\mathbf{1} - \rho}{\rho} \right] \quad (1.23)$$

This is done in DMFT_HRHO.

2. Construct the Greens function \hat{G}_ρ

$$\mathbf{G}_\rho(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu) \mathbf{1} - \mathbf{h}_\rho \right]^{-1} \quad (1.24)$$

3. For each site (or correlated cluster), extract the local part of the Green's function. Then transform Green's function and U-tensor to the local orthonormal basisset (such as local natural orbitals.) Furthermore expand U-tensor and Green's function into the spin-up spin-down spinor representation.
4. pass the local Green's function and the transformed U-tensor to the solver interface:

SOLVER

The external solver may do further approximations to the U-tensor. Then it calculates the non-Hartree-Fock contribution of the Luttinger Ward functional, i.e.

$$\Phi^{LW}[G, \hat{W}_2] - \Phi^{LW,HF}[G, \hat{W}_2] \quad (1.25)$$

and the non-Hartree-Fock contribution of the self energy

$$\Sigma_{2,a,b}(i\omega_\nu) = \Sigma_{a,b}(i\omega_\nu) - \Sigma_{a,b}^{HF}(i\omega_\nu) = \frac{\beta \delta \Phi^{LW}[G, \hat{W}_2]}{\delta G_{b,a}(i\omega_\nu)} - \frac{\beta \delta \Phi^{LW,HF}[G, \hat{W}_2]}{\delta G_{b,a}(i\omega_\nu)} \quad (1.26)$$

Later we should also calculate the derivative of the energy terms with respect to the local U-tensor. This derivative is the two-particle density matrix.

$$N_{a,b,c,d} = \frac{\beta \delta \Phi^{LW}[G, \hat{W}_2]}{\delta U_{a,b,c,d}} - \frac{\beta \delta \Phi^{LW,HF}[G, \hat{W}_2]}{\delta U_{a,b,c,d}} \quad (1.27)$$

5. convert the self energy and the derivative of the U-tensor back to the non-orthonormal set of local orbitals.
6. The new Green's function $\bar{\mathbf{G}}$ has the form

$$\bar{\mathbf{G}}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \mathbf{h}_p - \Sigma_2(i\omega_\nu) - \underbrace{(\mathbf{h}' - \mathbf{h}_p + \Sigma_1)}_{\mathbf{\Gamma}} \right]^{-1} \quad (1.28)$$

where the Lagrange multiplier $\mathbf{\Gamma}$ needs to be adjusted until the density matrix constraint

$$\boldsymbol{\rho} = \frac{1}{\beta} \sum_{\nu} e^{i\beta\hbar\omega_\nu 0^+} \mathbf{G}(i\omega_\nu) \quad (1.29)$$

is fulfilled.

For this purpose, we linearize the constraint equation in the Lagrange multiplier

$$\boldsymbol{\rho} = \frac{1}{\beta} \sum_{\nu} e^{i\beta\hbar\omega_\nu 0^+} \left[\bar{\mathbf{G}}(i\omega_\nu) + \bar{\mathbf{G}}(i\omega_\nu) \delta \mathbf{\Gamma} \bar{\mathbf{G}}(i\omega_\nu) \right] \quad (1.30)$$

Note that $\mathbf{\Gamma}$ is, like \mathbf{h}' , a non-local Hamiltonian, that in principle connects arbitrary local orbitals with each other. In a Bloch representation, $\mathbf{\Gamma}$ is a \mathbf{k} -dependent matrix, which connects all local orbitals in the unit cell with each other.

With the correct Lagrange multiplier, the new Green's function is obtained.

7. The new Green's function is feed back into the calculation of the Luttinger Ward functional and its self energy.

Energy contribution

When the loop is converged, we evaluate

$$\mathbf{h}' = \mathbf{h}_\rho + \mathbf{\Gamma} - \mathbf{\Sigma}_1 \quad (1.31)$$

and from it Eq. 1.21

$$\begin{aligned} Q_{dyn,\beta}^{\hat{W}}[\rho] = & \left\{ \Phi_{\beta}^{LW}[\mathbf{G}, \hat{W}] - \frac{1}{\beta} \sum_{\nu} \text{Tr} \left\{ \ln \left[\mathbf{1} - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' + \mathbf{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \right\} \right. \\ & + \left(\mathbf{h}' + \mathbf{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \mathbf{G}(i\omega_{\nu}) - \underbrace{\left[\mathbf{G}(i\omega_{\nu}) - \left(i\hbar\omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1}}_{=0} \left(\mathbf{h}' - \mathbf{h}_{\rho} \right) \left. \right\} \\ & - \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left[\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \right] \end{aligned} \quad (1.32)$$

1.4.2 Dahlen's trick

The evaluation fo the logarithm is problematic, because Green's function and self energy are not hermitean. In appendix B of their paper[5] Dahlen et al proposed a trick to solve the problem.

It is not yet clear to me if this trick is correct.

The trick rests on the assumption of the following identity

$$\text{Tr} \ln(\mathbf{A}\mathbf{A}^{\dagger}) \stackrel{?}{=} \text{Tr} \ln(\mathbf{A}) + \text{Tr} \ln(\mathbf{A}^{\dagger}) \quad (1.33)$$

which should be valid for arbitrary non-hermitean matrices \mathbf{A} .

Consider the singular value decompostion of $\text{mat} \mathbf{A} = \mathbf{U}\mathbf{\Sigma}\mathbf{V}^{\dagger}$, where \mathbf{U} and \mathbf{V} are unitary matrices and $\mathbf{\Sigma}$ is a diagonal matrix with real, non-negative numbers, the singular values, on the diagonal.

$$\begin{aligned} \text{Tr} \ln(\mathbf{A}\mathbf{A}^{\dagger}) &= \text{Tr} \ln(\mathbf{U}\mathbf{\Sigma}\mathbf{V}^{\dagger}\mathbf{V}\mathbf{\Sigma}^{\dagger}\mathbf{U}^{\dagger}) = \text{Tr} \ln(\mathbf{U}\mathbf{\Sigma}\mathbf{\Sigma}^{\dagger}\mathbf{U}^{\dagger}) = \text{Tr} \left[\mathbf{U} \ln(\mathbf{\Sigma}\mathbf{\Sigma}^{\dagger}) \mathbf{U}^{\dagger} \right] \\ &= \text{Tr} \left[\ln(\mathbf{\Sigma}\mathbf{\Sigma}^{\dagger}) \right] = \sum_i \ln(s_i) + \sum_i \ln(s_i) = \text{Tr} \ln[\mathbf{\Sigma}] + \text{Tr} \ln[\mathbf{\Sigma}^{\dagger}] \\ \text{Tr} \ln(\mathbf{A}) + \text{Tr} \ln(\mathbf{A}^{\dagger}) &= \text{Tr} \ln(\mathbf{U}\mathbf{\Sigma}\mathbf{V}^{\dagger}) + \text{Tr} \ln(\mathbf{V}\mathbf{\Sigma}^{\dagger}\mathbf{U}^{\dagger}) \end{aligned} \quad (1.34)$$

Unfortunately $\mathbf{V}^{\dagger}\mathbf{U} \neq \mathbf{1}$ so that we cannot simply convert the power series expansion into one of the singular values. **This is the problem.**

1.4.3 Traditional formulation of the dynamical mean-field theory

The loop of solver takes as input a local Greens function and a U-tensor. It produces the value of the Luttinger-Ward functional, and its derivatives of the Luttinger-Ward functional with respect to Greens function and U-tensor.

The algorithm is the following

1. choose a hybridization function $\Delta^{in}(i\omega_{\nu})$

2. calculate an output Greens function \mathbf{G}^{out} as follows¹: **Caution, I generalized Dieter's Equations without checking. Signs, factors, indices, etc. may be wrong!**

$$\begin{aligned}
 G_{\alpha,\beta}^{out}(i\omega_\nu) &:= -\frac{1}{Z} \int \prod_{\gamma} Dc_{\gamma}^* Dc_{\gamma} c_{\alpha}(i\omega_\nu) c_{\beta}^*(i\omega_\nu) e^{-S_{loc}} \\
 Z &= \int \prod_{\gamma} Dc_{\gamma}^* Dc_{\gamma} e^{-S_{loc}} \\
 S_{loc} &= -\int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \sum_{\alpha,\beta} c_{\alpha}^*(\tau_1) \left[i\hbar\omega_\nu \delta_{\alpha,\beta} - \Delta_{\alpha,\beta}^{in}(i\omega_\nu) \right] c_{\beta}(\tau_2) \\
 &\quad - \int_0^\beta d\tau \sum_{\alpha,\beta,\gamma,\delta} U_{\alpha,\beta,\delta,\gamma} c_{\alpha}^* c_{\beta}^* c_{\gamma} c_{\delta}
 \end{aligned}$$

Note, that only the hybridization function Δ^{in} and the U-tensor enter in this calculation. The only result is the output Greens function \mathbf{G}^{out} .

3. Convert the Green's function into a self energy Σ^{out} using Dyson's equation

$$\Sigma_{\alpha,\beta}^{out}(i\omega_\nu) := -\left(G^{out}(i\omega_\nu)\right)_{\alpha,\beta}^{-1} + i\hbar\omega_\nu \delta_{\alpha,\beta} - \Delta_{\alpha,\beta}^{in}(i\omega_\nu) \quad (1.35)$$

4. Use the Greens function \mathbf{G} passed to the solver as input argument (not \mathbf{G}^{out} !) to extract a new hybridization function Δ^{out}

$$\Delta_{\alpha,\beta}^{out}(i\omega_\nu) := -\left(G(i\omega_\nu)\right)_{\alpha,\beta}^{-1} + i\hbar\omega_\nu \delta_{\alpha,\beta} - \Sigma_{\alpha,\beta}^{out}(i\omega_\nu) \quad (1.36)$$

The steps given above yield a unique mapping from Δ^{in} to Δ^{out} . In other words the output hybridization function is a unique functional of the input hybridization function, the input Greens function and the U-tensor.

$$\Delta^{out} = F[\Delta^{in}, \mathbf{G}, \mathbf{U}] \quad (1.37)$$

The mapping depends on exactly two quantities, namely the input Green's function and the U-tensor. Self-consistency yields the fixed point $\Delta^{out} = \Delta^{in}$ of this mapping. At this fixed point, Eq. 1.36 and Eq. 1.35 yield

$$G_{\alpha,\beta}^{out}(i\omega_\nu) = G_{\alpha,\beta}(i\omega_\nu) \quad (1.38)$$

and

$$G_{\alpha,\beta}(i\omega_\nu) = \left[i\hbar\omega_\nu \mathbf{1} - \Sigma^{out}(i\omega_\nu) - \Delta^{in}(i\omega_\nu) \right]_{\alpha,\beta}^{-1}. \quad (1.39)$$

¹(D. Vollhardt *Dynamical Mean Field Theory for Strongly Correlated Materials in The LDA+DMFT Approach to Strongly Correlated Materials* E. Pavarini, E. Koch, D. Vollhardt and A. Lichtenstein Eds, Forschungszentrum Juelich 2011. Eq. 23.ff)

1.4.4 The interface

The Luttinger-Ward functional and its derivative, the self energy is obtained from an external solver. The solver is linked into the subroutine DMFT_SOLVERIO.

This routine supplies the local Green's function G and its first three Laurent expansion terms GLAUR, as well as the U-tensor U . All quantities are prepared with respect to a orthonormal one-particle basis². These orbitals are two-component spinor orbitals. The order of the orbitals is arbitrary, and there is not necessarily a division between spin-up and spin-down orbitals.

Note also, that the nature of the orbitals and also their order may change arbitrarily from one iteration to the next. However, the orbitals stay the same during the evaluation of a density matrix functional for a given density matrix ρ .

The routine expects back $\Delta\Phi^{LW} = \Phi^{LW} - \Phi^{LW,HF}$, the value of the Luttinger-Ward functional Φ^{LW} minus its Hartree-Fock value $\Phi^{LW,HF}$. The Hartree-Fock value of the Luttinger-Ward functional can be calculated directly from the density matrix given by Eq. 1.5 as

$$\Phi^{LW,HF} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} (\rho_{b,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b}) \quad (1.40)$$

Similarly, the self energy and its Laurent expansion terms follow from

$$\begin{aligned} \Delta\Sigma_{a,b}(i\omega_\nu) &= \beta \frac{\partial\Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \beta \frac{\partial\Phi^{LW,HF}}{\partial G_{b,a}(i\omega_\nu)} \\ &= \beta \frac{\partial\Phi^{LW}[\mathbf{G}, \hat{W}]}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d} (U_{a,c,b,d} - U_{a,c,d,b}) \rho_{c,d} \end{aligned} \quad (1.41)$$

1.5 Usage

1.5.1 Control file

A typical control file looks as follows. The DMFT object is activated by the block !NTBO with the value MODUS='DMFT'. The DMFT interface requires a finite temperature calculation, which is specified by the !MERMIN block, where the temperature is specified. A finite temperature calculation requires SAFEORTHO=T, so that the wave function dynamics converges to eigenstates of the Hamiltonian.

```
!CONTROL
!GENERIC NSTEP=500 DT=5. START=F !END
!FOURIER EPWPSI=30. CDUAL=2.0 !END
!DFT TYPE=10
!NTBO MODUS='DMFT' !END
!END
!PSIDYN STOP=T FRIC=.05 SAFEORTHO=F
!AUTO FRIC(-)=0.3 FACT(-)=0.97 FRIC(+)=0.3 FACT(+)=1.0 MINFRIC=0.05 !END
!END
!MERMIN T[K]=4000. ADIABATIC=T RETARD=10. !END
```

²The formerly non-orthogonal orbitals on a single site are orthogonalized with each other.

```
!end
!EOB
```

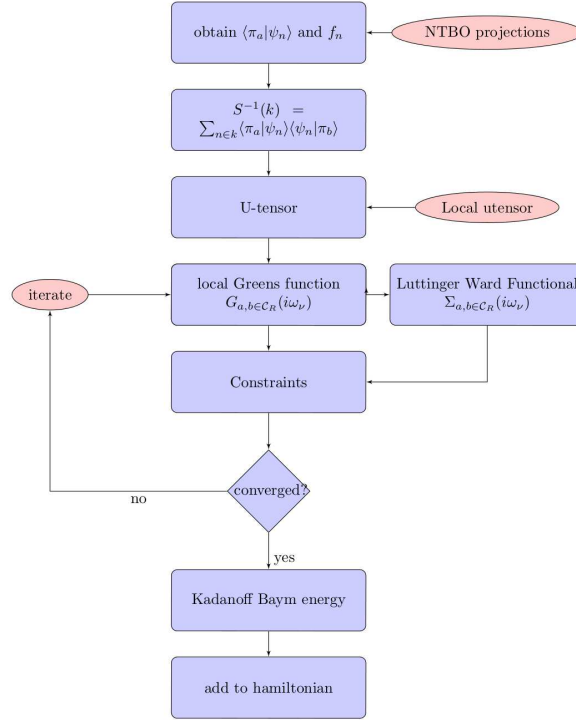
1.5.2 Structure file

A typical structure file may look as follows. New are the !NTBO subblocks.

```
!STRUCTURE
!GENERIC LUNIT[AA]= 3.8 EUNIT[EV]=T !END
!OCCUPATIONS EMPTY=10 NSPIN=2 SPIN[HBAR]=1.5 !END
!KPOINTS DIV=1 1 1 SHIFT=1 1 1 !END
!SPECIES NAME='CA' ID='CA_HBS_SC' NPRO=2 2 1
!NTBO NOFL=1 0 0 RAUG/RCOV=0.8 RTAIL/RCOV=1.4 TAILLAMBDA=2. 1.
CV=F FOCKSETUP=F !END
!END
!SPECIES NAME='MN' ID='MN_HBS' NPRO=1 1 1
!NTBO NOFL=1 0 1 RAUG/RCOV=1.2 RTAIL/RCOV=1.4 TAILLAMBDA=2. 1.
LHFWeight=0.25 CV=F FOCKSETUP=F !END
!END
!SPECIES NAME='O_' ID='O_.75_6.0' NPRO=1 1 0
!NTBO NOFL=1 1 RAUG/RCOV=1.2 RTAIL/RCOV=1.4 TAILLAMBDA=2. 1.
CV=F FOCKSETUP=F !END
!END
!LATTICE T= 1.0000 0.0000 0.0000000000
0.0000 1.0000 0.0000000000
0.0000 0.0000 1.0000000000 !END
!ATOM NAME= 'CA1' R= 0.0 0.0 0.0 !END
!ATOM NAME= 'MN1' R= 0.5 0.5 0.5 !END
!ATOM NAME= 'O_1' R= 0.0 0.5 0.5 !END
!ATOM NAME= 'O_2' R= 0.5 0.0 0.5 !END
!ATOM NAME= 'O_3' R= 0.5 0.5 0.0 !END
!ORBPOT_X
!POT ATOM='MN(UP)1' VALUE=+.2 TYPE='D' RC=1.5 S=1 !END
!END
!END
!EOB
```

1.6 Description of Subroutines

1.6.1 Flowchart



1.6.2 Data exchange of the Object wit the outer world

1.6.3 DMFT\$GREEN

DMFT\$GREEN is the main subroutine of the DMFT object. It is called from the LMTO object, which also provides the projections onto the tight-binding orbitals.

```

call dmft_ini()
call dmft_collecthamiltonian()
call dmft_collectfulldenmat()
call dmft_utensor()
call dmft_smat()
call dmft_natorb()
call dmft_hrho()
call dmft_constraints('hrho')
do iter=1,3
  call dmft_gloc_withatomset()
  call dmft_solver(etot)
  call dmft_constraints('h0')
enddo ! end of loop over iterations to enforce constraint
call dmft_detot(svar)
etot=etot+svar

```

```

call energylist$set('dmft interface',etot)
call energylist$add('local correlation',etot)
call energylist$add('total energy',etot)
call dmft_addtohpsi()

```

1. DMFT_COLLECTHAMILTONIAN:

$$\langle \pi_a | \psi_n \rangle \quad (1.42)$$

$$\rho_{a,b}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle f_n(\vec{k}) \langle \psi_n(\vec{k}) | \pi_b \rangle \quad (1.43)$$

2. DMFT_COLLECTFULLDENMAT: Calculates the local density matrix, but for all orbitals on each atom. This is needed for the double counting term.

3. DMFT_UTENSOR:

$$U_{a,b,c,d} = \alpha \int d^4x \int d^4x' \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x}') \chi_c(\vec{x}) \chi_d(\vec{x}')}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \quad (1.44)$$

Only the U-tensor for equal spin-electrons is stored. α is a scale factor that mimics the screening of the U-tensor. It is called LHFWEIGHT.

4. DMFT_SMAT:

$$\mathbf{S}^{-1}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle \langle \psi_n(\vec{k}) | \pi_b \rangle \quad (1.45)$$

Note that the inverse overlap matrix is spin and orbital dependent. $\mathbf{S}(\vec{k})$ and $\mathbf{S}^{-1}(\vec{k})$ are put on the KSET structure.

5. DMFT_NATORB: Construct a set of local, orthonormal orbitals $|\phi\rangle$.

Ther atomicn natural orbitals are the eigenstates o9f, which are defined by

$$\sum_{a,b} \langle \phi_i | \pi_a \rangle (\mathbf{S}_R)_{a,b} \langle \pi_b | \phi_j \rangle = \delta_{i,j} \quad (1.46)$$

where \mathbf{S}_R is the local overlap operator obtained from projection of $\sum_{\vec{k}} w_{\vec{k}} \mathbf{S}^{-1}(\vec{k})$ onto the local orbitals and inversion of the projection matrix.

Construct transformation matrices $\langle \chi | \phi \rangle$ and $\langle \pi | \phi \rangle$ onto an orthonormal local basisset $|\phi\rangle$, so that

$$\sum_{a,b} \langle \phi_i | \pi_a \rangle (\mathbf{S}_R)_{a,b} \langle \pi_b | \phi_j \rangle = \delta_{i,j} \quad (1.47)$$

where \mathbf{S}_R is the local overlap operator obtained from projection of $\sum_{\vec{k}} w_{\vec{k}} \mathbf{S}^{-1}(\vec{k})$ onto the local orbitals and inversion of the projection matrix.

The two matrices are related by

$$\langle \pi | \phi \rangle = \mathbf{S}_R^{-1} \langle \pi | \phi \rangle \quad (1.48)$$

Alternatively, (using a hard-wired switch) one can select a transformation onto natural orbitals, that is a representation for which also the local density matrix is diagonal.

$$\rho_R \langle \chi | \phi \rangle = \mathbf{S}_R^{-1} \langle \chi | \phi \rangle \bar{\chi} \quad (1.49)$$

The transformation matrices are stored in the fully non-collinear data model in `atomset%natorb%piphi` and `atomset%natorb%chphi`.

6. DMFT_HRHO: For each k-point solve the generalized eigenvalue problem

$$\rho(k) \mathbf{V}(\vec{k}) = \mathbf{S}^{-1}(\vec{k}) \mathbf{V}(\vec{k}) \mathbf{f}(\vec{k})$$

where the \mathbf{f}_k is a matrix that contains k-dependent occupations, which differs from the occupations f_n .

$$\mathbf{h}_\rho(\vec{k}) - \mu = \mathbf{V}^{\dagger,-1}(\vec{k}) k_B \ln \left[\frac{\mathbf{1} - \mathbf{V}^\dagger(\vec{k}) \rho(\vec{k}) \mathbf{V}(\vec{k})}{\mathbf{V}^\dagger(\vec{k}) \rho(\vec{k}) \mathbf{V}(\vec{k})} \right] \mathbf{V}^{-1}(\vec{k}) \quad (1.50)$$

\mathbf{h}_ρ is kept as `KSET%HRHO`.

7. DMFT_CONSTRAINTS: Determines $\mathbf{h}'(\vec{k})$ such that

$$\rho(\vec{k}) = k_B T \sum_{\nu} \left[(i\omega_{\nu} + \mu) \mathbf{S}(\vec{k}) - \mathbf{h}' - \boldsymbol{\Sigma}(i\omega_{\nu}) \right]^{-1} \quad (1.51)$$

\mathbf{h}' is kept as `KSET%H0`.

8. DMFT_GLOC: The local Green's function is obtained as Brillouin zone integral over the k-dependent greens function after projecting onto the correlated orbitals on the specified site.

$$\mathbf{G}_R(i\omega_{\nu}) = \sum_k w(\vec{k}) \left[(i\omega_{\nu} + \mu) \mathbf{S}(\vec{k}) - \mathbf{h}' - \boldsymbol{\Sigma}(i\omega_{\nu}) \right]_R^{-1} \quad (1.52)$$

The result and its Laurent expansion terms are stored in `atomset%gloc` and `atomset%gloc Laur`.

9. DMFT_SOLVER: Calculates the self energy for the local Green's functions. The Hartree-Fock term is separated out and calculated directly. Only the dynamic contribution is taken from an external solver. The double counting term is calculated as in the LMTO module. **For the double counting in the HF approximation only the exchange part is removed, while here also the correlation contribution should be taken out, because that is explicitly added.**

The Hartree-Fock contribution is added directly to the double counting term. Thus the self energy `atomset%sigma` is only the dynamical contribution.

Green's function and U-tensor are transformed into the representation of orthonormal local orbitals, and the self-energy is transformed back accordingly.

The external solver shall evaluate

$$\Phi^{LW}[\mathbf{G}(i\omega_{\nu}), \hat{W}] - \Phi^{LW,HF}[\mathbf{G}(i\omega_{\nu}), \hat{W}] \quad (1.53)$$

where

$$\begin{aligned}\Phi^{LW,HF}[\mathbf{G}, \hat{W}] &= \frac{1}{2} \frac{1}{\beta^2} \sum_{\nu, \nu'} e^{i\beta \hbar \omega_{\nu} 0^+} e^{i\beta \hbar \omega_{\nu'} 0^+} \sum_{a,b,c,d} U_{a,b,d,c} \\ &\quad \times \left(G_{d,a}(i\omega_{\nu}) G_{c,b}(i\omega_{\nu'}) - G_{c,a}(i\omega_{\nu}) G_{d,b}(i\omega_{\nu'}) \right) \\ &= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left(\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \right)\end{aligned}\quad (1.54)$$

Correspondingly, the self energy to be returned must have the Hartree-Fock term subtracted out. That is, the double counting term must be the Hartree-Fock term.

$$\Sigma_{a,b}(i\omega_{\nu}) = \frac{\beta \partial \Phi^{LW}}{\partial G_{b,a}(i\omega_{\nu})} - \sum_{c,d} \left(U_{a,c,b,d} - U_{a,c,d,b} \right) \rho_{d,c}$$

The external solver may make approximations to the U-tensor or to the Kadanoff-Baym functional. However, they need to be consistent for Luttinger-Ward functional and the corresponding Hartree-Fock term. Thus, it is ensured that there is no approximation made beyond the dynamic effects.

10. DMFT_DETOT: Adds the non-local contribution to the total energy

$$\begin{aligned}-\frac{1}{\beta} \sum_{\nu} \text{Tr} \left\{ \ln \left[\mathbf{1} - \left(i\hbar \omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' + \boldsymbol{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \right\} \\ + \left(\mathbf{h}' + \boldsymbol{\Sigma}(i\omega_{\nu}) - \mathbf{h}_{\rho} \right) \mathbf{G}(i\omega_{\nu}) - \left[\mathbf{G}(i\omega_{\nu}) - \left(i\hbar \omega_{\nu} + \mu \right) \mathbf{1} - \mathbf{h}_{\rho} \right]^{-1} \left(\mathbf{h}' - \mathbf{h}_{\rho} \right) \end{aligned} \quad (1.55)$$

- **At this point I am still using the Dahlen trick.**
- **Is it correct that only the dynamic self energy is used?**

11. DMFT_ADDTOHPSI: Adds the energy derivative to `this%htbc` of the `waves_module`. There are two contributions, a k-dependent term which is restricted to the correlated orbitals and an site-dependent term that acts on all local orbitals.

- The site-specific term contains the double counting term for the DFT functional and the Hartree-Fock contribution to the self energy.

1.6.4 DMFT_INI

Hardwired data

- NOMEGA (N_{ω})

Inherited data

- NDIM from `waves_module`
- NSPIN from `waves_module`
- NKPTL from `waves_module`

- NAT from ATOMLIST Object
- isp(IAT)
- WKPT and (NKPT,KMAP) from DYNOC Object
- KBT from `dynocc$getr8a('temp')` from `paw_occupations.f90`
- `atomset(iat)%lhweight` from `lmto_module`

Derived data

- NDIMD (=1 for NDIM=1,NSPIN=1; =2 for NDIM=1,NSPIN=2; =4 for NDIM=2,NSPIN=1)
- List of positive Matsubara frequencies

$$\omega_\nu = (2\nu - 1)\pi k_B T \quad \text{for } \nu = 1, \dots, N_\omega \quad (1.56)$$

- Atomset structure

`atomset\%nloc` number of correlated orbits on this atom
`atomset\%ICHI1` first value of ICHI index for this atom
`atomset\%ICHI2` last value of ICHI index for this atom

- KSET structure

`KSET\%WKPT` geometric k-point weight

1.6.5 DMFT_UTENSOR

Obtain the onsite Utensor for all local orbitals from subroutine `dmft_ulocal` reduces it to the entries for the correlated orbitals.

In `DMFT_ULOCAL` it obtains the local U-tensor from the `POTPAR%TAILED%U` and `SBAR` of the `LMTO` Object. This object contains the extended ϕ and $\dot{\phi}$ functions

$$|\chi_a\rangle = |\phi_a\rangle - \sum_{b; R_b=R_a} |\dot{\phi}_b\rangle \bar{S}_{a,b} \quad (1.57)$$

The U-tensor is constructed in `LMTO_MAKETAILEDPARTIALWAVES`. `LMTO_ULITTLE` constructs

$$u_{\ell,a,b,c,d} = \frac{2\ell+1}{4\pi} \int dr r^2 R_c(r) R_d(r) \left[\int d^3 r' V_\ell(r, r') R_a(r') R_b(r') \right] \quad (1.58)$$

where the kernel $V_\ell(r, r')$ is the one used by `RADIAL$POISSON`. **There is a input parameter that determines which ℓ values are considered.**

In `LMTO_UTENSOR` the U-tensor elements are composed according to

$$U_{a,b,c,d} = \sum_{\ell} \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} u_{\ell,b,d,c,a} C_{L,L_b,L_d} C_{L,L_c,L_a} \quad (1.59)$$

The U-tensor is screened by a factor LHFWEIGHT. It is obtained from the `lmtto_module` either as the global value HFWEIGHT or the individual value from hybridsetting. This very same factor is used for the double counting term.

Thus the U-tensor is defined as³ in Eq. 1.44, i.e. as

$$U_{a,b,c,d} = \int d^4x \int d^4x' \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x}') \chi_c(\vec{x}) \chi_d(\vec{x}')}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \quad (1.60)$$

The U-tensor contributes only if $\sigma_a = \sigma_c$ and if $\sigma_b = \sigma_d$. With this definition the Hartree and exchange energy have the form

$$\begin{aligned} E_H &= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \rho_{c,a} \rho_{d,b} \\ E_X &= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \rho_{c,b} \rho_{d,a} \end{aligned} \quad (1.61)$$

1.6.6 DMFT_NATORB

Construct natural orbitals in the space of correlated orbitals on one site.

For this purpose, I redefine the unity operator for the sub Hilbert space of correlated orbitals.

$$\hat{1}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle \langle \pi_\alpha | \psi_n \rangle \langle \psi_n | \pi_\beta \rangle \langle \chi_\beta| \quad (1.62)$$

which allows one to identify the inverse overlap matrix in this sub-Hilbert space.

$$\mathbf{S}_{\alpha,\beta}^{-1} = \sum_{\alpha,\beta} \sum_n \langle \pi_\alpha | \psi_n \rangle \langle \psi_n | \pi_\beta \rangle \quad (1.63)$$

Note, however, that this matrix lives only on a single site! We can obtain it from the k-dependent \mathbf{S}^{-1} by summing over k-points and cutting out the corresponding submatrix. However, it is not allowed to do the same thing for the overlap matrix itself!

The one-particle density matrix in this sub-Hilbert space has the form

$$\hat{\rho}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle \langle \pi_\alpha | \psi_n \rangle f_n \langle \psi_n | \pi_\beta \rangle \langle \chi_\beta| \quad (1.64)$$

The eigenvalue equation

$$\sum_{\alpha,\beta} |\chi_\alpha\rangle \langle \pi_\alpha | \hat{\rho} | \pi_\beta \rangle \langle \chi_\beta | \phi_j \rangle = \sum_{\alpha,\beta} |\chi_\alpha\rangle \langle \pi_\alpha | \pi_\beta \rangle \langle \chi_\beta | \phi_j \rangle f_j = \sum_{\alpha,\beta} |\chi_\alpha\rangle \mathbf{S}_{\alpha,\beta}^{-1} \langle \chi_\beta | \phi_j \rangle \bar{f}_j \quad (1.65)$$

provides us with a new set of occupations \bar{f} of local natural orbitals and eigenvectors $\langle \chi_\beta | \phi_j \rangle = V_{\beta,j}$.

$$\rho \mathbf{V} = \mathbf{S}^{-1} \mathbf{V} \bar{f} \quad \text{with} \quad \mathbf{V}^\dagger \mathbf{S}^{-1} \mathbf{V} = \mathbf{1} \quad (1.66)$$

³ $\vec{x} = (\vec{r}, \sigma)$

The natural orbitals are

$$|\phi_j\rangle = \sum_{\alpha, \beta \in \mathcal{C}_R} |\chi_\alpha\rangle \underbrace{\mathbf{S}_{\alpha, \beta}^{-1} \langle \chi_\beta | \phi_j \rangle}_{\mathbf{1}_{\mathcal{C}_R}} = \sum_{\alpha \in \mathcal{C}_R} |\chi_\alpha\rangle \underbrace{(\mathbf{S}^{-1} \mathbf{V})_{\alpha, j}}_{\mathbf{V}_{\alpha, j}^{\dagger, -1}} \quad (1.67)$$

The matrix \mathbf{V} is placed into the structure `atomset%natorb%chiphi`, together with the occupations \bar{f} which are in `atomset%natorb%f`. In addition we store $\mathbf{S}^{-1} \mathbf{V} = \mathbf{V}^{\dagger, -1}$ in `atomset%natorb%piphi`. All data in `atomset%natorb` are stored in the fully non-collinear model.

1.6.7 DMFT_HRHO

The conversion of occupations into energies is done in the routine `DMFT_EOFF`.

$$\epsilon = k_B \begin{cases} \ln\left(\frac{1-f}{f}\right) & \text{for } f_0 < f < 1 - f_0 \\ a + b(f - f_0) & \text{for } f < f_0 \\ -a + b(f - (1 - f_0)) & \text{for } f > 1 - f_0 \end{cases} \quad (1.68)$$

where a and b are determined such that the mapping is differentiable. The value of f_0 is currently set to 10^{-6} .

1.6.8 DMFT_CONSTRAINTS

The two equations are solved iteratively

$$\bar{\rho}(\vec{k}) = k_B T \sum_{\nu} \underbrace{\left[(i\omega_{\nu} + \mu) \mathbf{S}(\vec{k}) - \mathbf{h}(\vec{k}) - \sum_R \Sigma_R(i\omega_{\nu}) \right]}_{\mathbf{G}(\vec{k})}^{-1} \quad (1.69)$$

$$\delta \mathbf{h} = 4k_B T \mathbf{S}(\vec{k}) \left[\bar{\rho}(\vec{k}) - \rho(\vec{k}) \right] \mathbf{S}(\vec{k}) \quad (1.70)$$

This equation is a Newton scheme with the Green's function in the slope estimate replaced by the non-interacting Green's function with zero potential self energy and chemical potential. **Should there be convergence problems, it may be improved by using the static Green's function \mathbf{G}_{ρ} instead.**

The Laurent expansion terms are

$$\begin{aligned} \mathbf{G}^{(1)}(\vec{k}) &= \mathbf{S}^{-1}(\vec{k}) \\ \mathbf{G}^{(2)}(\vec{k}) &= \mathbf{S}^{-1}(\vec{k}) \left(-\mu \mathbf{S}(\vec{k}) + \mathbf{h}'(\vec{k}) + \Sigma^{(1)}(\vec{k}) \right) \mathbf{S}^{-1}(\vec{k}) \\ \mathbf{G}^{(2)}(\vec{k}) &= \mathbf{S}^{-1}(\vec{k}) \left[\mathbf{S}^{-1}(\vec{k}) \left(-\mu \mathbf{S}(\vec{k}) + \mathbf{h}'(\vec{k}) + \Sigma^{(1)}(\vec{k}) \right) \mathbf{S}^{-1}(\vec{k}) + \Sigma^{(2)}(\vec{k}) \right] \mathbf{S}^{-1}(\vec{k}) \end{aligned} \quad (1.71)$$

1.6.9 DMFT_SOLVER

The solver first calculates the Hartree-Fock contribution to the Luttinger-Ward functional as defined in Eq. 1.40 and the corresponding self energy. Because the Hartree-Fock self energy is

static and because it does not affect the density matrix constraint, it is added together with the double counting correction to `atomset%denmat%h`.

Then the double counting correction is calculated. **At the moment we still subtract out only the exchange part, which is consistent with the hybrid functionals but not as double counting for correlation.**

Finally the data are prepared for the solver in `DMFT_dynamicsolver`. The `dynamicsolver` transforms all data into a spin-orbital representation of orthonormal states, which may be either natural orbitals derived from the local density matrix or which may be eigenstates of the local overlap matrix.

Appendix A

Matsubara frequencies

The Matsubara frequencies for Fermions are

$$\omega_\nu = (2\nu + 1) \frac{\pi}{\hbar\beta}$$

A Matsubara sum has the general form

$$\frac{1}{\beta} \sum_{\nu \in \mathbb{Z}} g(i\omega_\nu)$$

where $g(i\omega_\nu)$ is a analytic function.

A.1 Evaluation using residual theorem

In order to evaluate the Matsubara sum we introduce a **Matsubara weighting functions**, that has simple poles at $i\omega_\nu$, so that we can exploit the residuum theorem. For Fermions, two such weighting functions are used

$$h^\pm(z) = \frac{\mp\beta\hbar}{1 + e^{\pm\beta\hbar z}} \quad (\text{A.1})$$

The poles of the Matsubara weighting functions obey

$$1 + e^{\pm\beta\hbar z} = 0 \quad (\text{A.2})$$

With the help of

$$e^{\pm\beta\hbar z} = e^{\pm\beta\hbar \text{Re}(z)} e^{\pm i\beta\hbar \text{Im}(z)} = e^{\beta\hbar \text{Re}(z)} \cos(\beta\hbar \text{Im}(z)) \pm i e^{\beta\hbar \text{Re}(z)} \sin(\beta\hbar \text{Im}(z))$$

Eq. A.2 can be rewrfitten in the form

$$\Rightarrow e^{\pm\beta\hbar \text{Re}(z)} \cos(\beta\hbar \text{Im}(z)) = -1 \quad \text{and} \quad e^{\pm\beta\hbar \text{Re}(z)} \sin(\beta\hbar \text{Im}(z)) = 0 \quad (\text{A.3})$$

The second equation is obeyed for

$$\text{Im}(z) = \frac{n\pi}{\hbar\beta} \quad \text{for arbitrary integer } n \quad (\text{A.4})$$

The first equation yields

$$\begin{aligned} -1 &= e^{\pm\beta\hbar\text{Re}(z)} \cos(\beta\hbar\text{Im}(z)) = e^{\pm\beta\hbar\text{Re}(z)} \cos(n\pi) = e^{\pm\beta\hbar\text{Re}(z)} (-1)^n \\ \Rightarrow n-1 &= 2\nu \quad \text{for arbitrary integer } \nu, \text{ and } \text{Re}(z) = 0 \\ \Rightarrow n &= 2\nu + 1 \end{aligned} \quad (\text{A.5})$$

Thus we obtain that the poles of the Matsubara weighting function lie at the Matsubara frequencies.

$$z_\nu = i(2\nu + 1) \frac{\pi}{\hbar\beta} = i\omega_\nu \quad (\text{A.6})$$

Next we need to show that the weighting function at the poles behaves like $\frac{1}{z-z_0} + O(|z-z_0|^2)$, where z_0 is the position of the pole. For this purpose we expand the inverse

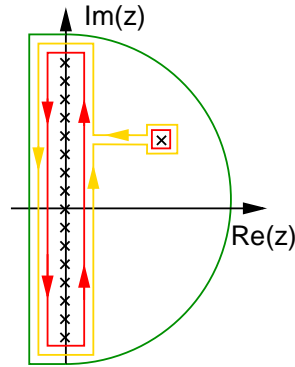
$$\frac{1}{\mp\hbar\beta} (1 + e^{\pm\hbar\beta z}) = \frac{1}{\mp\hbar\beta} \left(\underbrace{1 + e^{\pm\hbar\beta z_0}}_{=0} \pm \hbar\beta \underbrace{e^{\pm\hbar\beta z_0}}_{-1} (z - z_0) + O(|z - z_0|^2) \right) \quad (\text{A.7})$$

$$= (z - z_0) + O(|z - z_0|^2) \quad (\text{A.8})$$

With the help of the Matsubara weighting function we can evaluate the Matsubara sum as¹

$$\frac{1}{\beta} \sum_{n \in \mathbb{Z}} g(i\omega_n) = \frac{1}{2\pi i \beta} \oint g(z) h^\pm(z) = -\frac{1}{\beta} \sum_{z_0 \in \text{poles of } g} \text{Res}(gh^\pm, z_0) \quad (\text{A.9})$$

The minus sign occurs because the counter clockwise integration of the contour in the half plane turns into a clock-wise integration about the pole.



The integration is closed in the half plane with $\pm \text{Re}(z) > 0$, because there the corresponding weighting function h^\pm decays exponentially for $\text{Re}(z) \rightarrow \pm\infty$.

¹Cauchy's Integral formula:

$$\text{Res}(f, z_0) = \frac{1}{2\pi i} \oint_\gamma dz f(z)$$

where γ is a counter-clockwise contour around a pole z_0 of a function f , that can be expanded into a Laurent series about z_0 . The Residue is the prefactor of the term $1/(z - z_0)$ in the Laurent expansion.

A.1.1 Matsubara sums

From http://en.wikipedia.org/wiki/Matsubara_frequency we obtain the following expression for the **fermionic** summations. In these expressions the Matsubara frequencies are²

$$\omega_\nu = (2\nu + 1)\pi/(\hbar\beta) \quad (\text{A.10})$$

$$-k_B T \sum_\nu \ln[-i\hbar\omega_\nu + \epsilon] e^{i\omega_\nu 0^+} = -k_B T \ln[1 + e^{-\beta\epsilon}] \quad (\text{A.11})$$

$$k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)} e^{i\omega_\nu 0^+} = (1 + e^{\beta\epsilon})^{-1} \quad (\text{A.12})$$

$$k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)^n} = \frac{1}{(k_B T)^{n-1} (n-1)!} \partial_\epsilon^{n-1} \Big|_{x=\beta\epsilon} (1 + e^x)^{-1} \quad (\text{A.13})$$

j	1	2	3	4	5	6	7	8	9	10
$\partial_x^{j-1}(1 + e^x)^{-1}$	$\frac{1}{2}$	$-\frac{1}{4}$	0	$+\frac{1}{8}$	0	$-\frac{1}{4}$	0	$\frac{17}{16}$	0	$-\frac{31}{4}$
$\partial_x^{j-1}(1 + e^x)^{-1}$	0.5	-0.25	0	+0.125	0	-0.25	0	+1.0625	0	-7.75
j	11	12	13	14	15	16	17	18	19	20
$\partial_x^{j-1}(1 + e^x)^{-1}$	0	$\frac{691}{8}$	0	$-\frac{5461}{4}$	0	$\frac{929569}{32}$	0	$-\frac{3202291}{4}$	0	$+\frac{221930581}{8}$
$\partial_x^{j-1}(1 + e^x)^{-1}$	0	86.375	0	-1365.25	0	29.0×10^3	0	-800×10^3	0	27.7×10^6

A.1.2 Matsubara sums on finite grids

The finite Matsubara sums result in a Fermi function that does not reach zero or one. In particular, in the limit of ϵ level, that lies far from the chemical potential, the finite Matsubara sum falls off to $\frac{1}{2}$.

²They differ from those of the source by a factor $1/\hbar$.

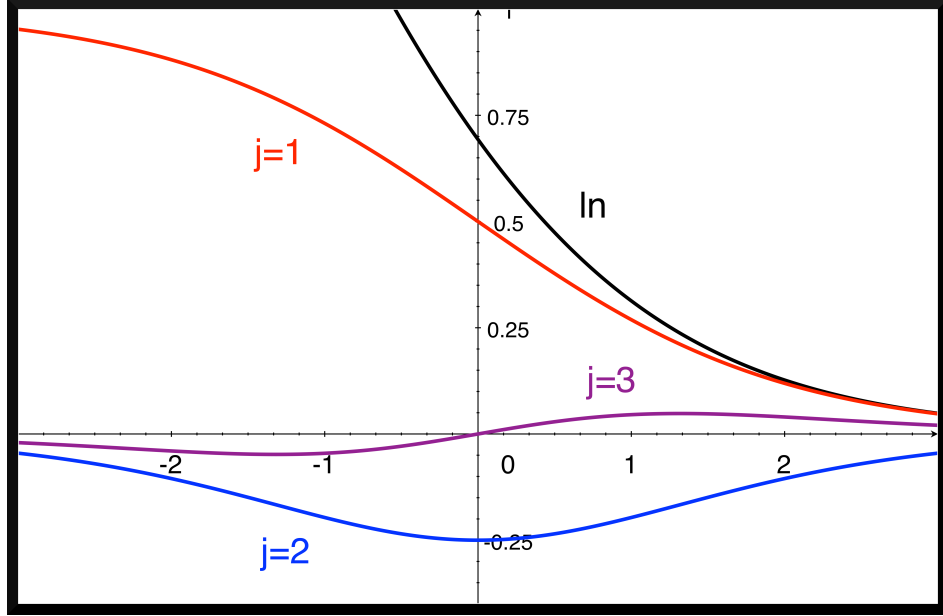


Fig. A.1: The result of Matsubara sums $-k_B T \sum_{\nu} \ln[-i\hbar\omega_{\nu} + \epsilon]$, $k_B T \sum_{\nu} \frac{1}{(i\hbar\omega_{\nu} - \epsilon)} e^{i\omega_{\nu} 0^+}$, $k_B T \sum_{\nu} \frac{1}{(i\hbar\omega_{\nu} - \epsilon)^2}$ as function of energy ϵ .

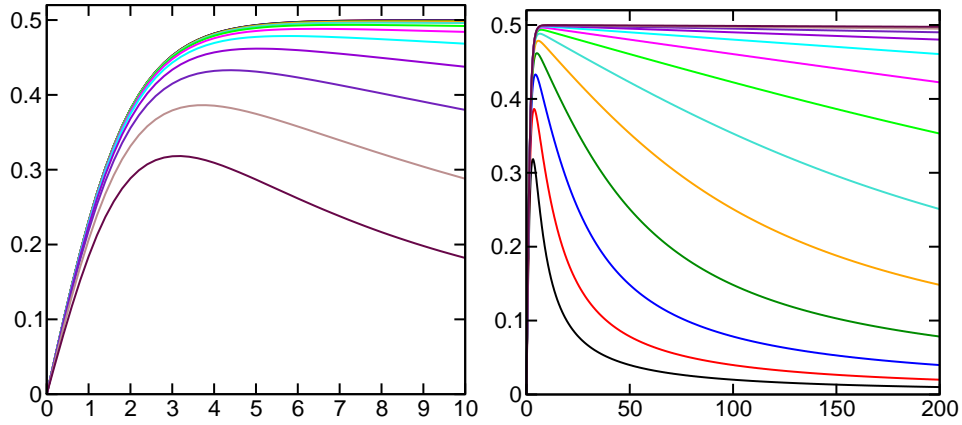


Fig. A.2: Fermi function calculated from a finite Matsubara sum. The function is shifted by $-\frac{1}{2}$ because no regularization has been done. The sums have been performed with 2^n grid points with $n = 0, \dots, 12$ for $k_B T = 1$.

Appendix B

The Green function

We construct the **lattice Green's function** defined as

$$\begin{aligned}\hat{G}^{lat}(i\omega_\nu) &= \left[(i\hbar\omega_\nu + \mu)\hat{1} - \hat{h} - \Delta\hat{\Sigma}(i\omega_\nu) \right]^{-1} \\ &\approx \sum_{n,n'} |\psi_n\rangle \underbrace{\left[\langle\psi_{n'}|(i\hbar\omega_\nu + \mu)\hat{1} - \hat{h} - \Delta\hat{\Sigma}(i\omega_\nu)|\psi_n\rangle \right]_{n,n'}^{-1}}_{G_{n,n'}^{lat}} \langle\psi_{n'}| \end{aligned} \quad (\text{B.1})$$

This expression is approximate, if the set of band states is not complete. In practice, corrections are required.

In the representation of the band states, the lattice Green's function has the form

$$G_{n,n'}^{lat}(i\omega_\nu) = \left[\langle\psi_{n'}|(i\hbar\omega_\nu + \mu)\hat{1} - \hat{h} - \Delta\hat{\Sigma}(i\omega_\nu)|\psi_n\rangle \right]_{n,n'}^{-1} \quad (\text{B.2})$$

Note, that the matrix to be inverted is not necessarily hermitean!

B.1 Generic properties

B.1.1 Green's function with negative Matsubara frequencies

Green's functions and self energies obey the relation

$$\mathbf{G}(-i\omega_\nu) = \mathbf{G}^\dagger(i\omega)$$

This allows one to store only half of the Green's functions.

Proof:

$$G_{\alpha,\beta}(i\omega_n) = -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{i\omega_n\tau} \langle \mathcal{T} \hat{c}_\alpha(\tau) \hat{c}_\beta^\dagger(0) \rangle \quad (\text{B.3})$$

$$\begin{aligned} \Rightarrow G_{\alpha,\beta}(-i\omega_n) &= -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{-i\omega_n\tau} \langle \mathcal{T} \hat{c}_\alpha(\tau) \hat{c}_\beta^\dagger(0) \rangle \\ &= \left\{ -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{i\omega_n\tau} \left\langle \left(e^{\beta(\hat{H}-\mu\hat{N})} \hat{c}_\alpha e^{-\beta(\hat{H}-\mu\hat{N})} \hat{c}_\beta^\dagger \right)^\dagger \right\rangle \right\}^* \\ &= \left\{ -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{i\omega_n\tau} \left\langle \hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})} \hat{c}_\alpha^\dagger e^{\beta(\hat{H}-\mu\hat{N})} \right\rangle \right\}^* \\ &\stackrel{cycl, perm}{=} \left\{ -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{i\omega_n\tau} \left\langle e^{\beta(\hat{H}-\mu\hat{N})} \hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})} \hat{c}_\alpha^\dagger \right\rangle \right\}^* \\ &= \left\{ -\frac{1}{\hbar} \int_0^{\hbar\beta} d\tau e^{i\omega_n\tau} \langle \mathcal{T} \hat{c}_\beta(\tau) \hat{c}_\alpha^\dagger \rangle \right\}^* \\ &= G_{\beta,\alpha}^*(i\omega_n) \end{aligned} \quad (\text{B.4})$$

B.1.2 Density of states

For a system of independent electrons, the number of particles is related to the chemical potential via

$$N(\mu) = \int_{-\infty}^{\infty} d\epsilon f_\mu(\epsilon) D(\epsilon), \quad (\text{B.5})$$

where D is the density of states. $f_\mu(\epsilon)$ is the Fermi function.

$$\frac{dN}{d\mu} = \int_{-\infty}^{\infty} d\epsilon \frac{\partial f_\mu(\epsilon)}{\partial \mu} D(\epsilon) = \underbrace{\int_{-\infty}^{\infty} d\epsilon \frac{\beta}{\cosh^2\left(\frac{1}{2}\beta(\epsilon - \mu)\right)} D(\epsilon)}_{\tilde{D}(\mu)} \quad (\text{B.6})$$

Thus $dN/d\mu$ provides us with a broadened density of states.

No we generalize the relation obtained for independent Fermions to interacting Fermions. The number of states function $N_{a,b}(\mu)$ can be obtained from the Green's function

$$N_{a,b}(\mu) = \frac{1}{\beta} \sum_{\nu} G_{a,b}(i\omega_{\nu}, \mu) e^{i\omega_{\nu}0^+} = \frac{1}{\beta} \sum_{\nu} \left[\left(\mathbf{G}(i\omega_{\nu}, \mu_0) \right)^{-1} + (\mu - \mu_0) \right]_{a,b}^{-1} e^{i\omega_{\nu}0^+} \quad (\text{B.7})$$

The smoothened density of states $\tilde{D}_{a,b}(\mu)$ is then obtained as derivative with respect to μ .

$$\begin{aligned} \tilde{D}_{a,b}(\mu) &\stackrel{\text{def}}{=} \frac{\partial N_{a,b}}{\partial \mu} \\ &= -\frac{1}{\beta} \sum_{\nu} \sum_c \left[\left(\mathbf{G}(i\omega_{\nu}, \mu_0) \right)^{-1} + (\mu - \mu_0) \mathbf{1} \right]_{a,c}^{-1} \left[\left(\mathbf{G}(i\omega_{\nu}, \mu_0) \right)^{-1} + (\mu - \mu_0) \mathbf{1} \right]_{c,b}^{-1} \end{aligned} \quad (\text{B.8})$$

B.2 Project Green's function onto local orbitals

For a Hamiltonian $\hat{h} = \sum_{a,b} |\pi_a\rangle h_{a,b} \langle \pi_b|$ and a self energy $\hat{\Sigma}(i\omega_\nu) = \sum_{a,b} |\pi_a\rangle \Sigma_{a,b}(i\omega_\nu) \langle \pi_b|$ acting on the local orbitals, the Green's function can be expressed as

$$\begin{aligned} \hat{G}(i\omega_\nu) = \sum_{a,b} |\chi_a\rangle \left\{ \sum_n \langle \pi_a | \psi_n \rangle \left[(i\hbar\omega_\nu + \mu) \delta_{n,n'} \right. \right. \\ \left. \left. - \sum_{c,d} \langle \psi_n | \pi_c \rangle (h_{c,d} + \Sigma_{c,d}(i\omega_\nu)) \langle \pi_d | \psi_{n'} \rangle \right]_{n,n'}^{-1} \langle \psi_{n'} | \pi_b \rangle \right\} \langle \chi_b | \end{aligned} \quad (\text{B.9})$$

We converted Eq. (B.1) into a basis of local orbitals $|\chi_a\rangle$ using $|\psi_n\rangle = \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle$.

In order to evaluate Green's function, we still need to refer to the band states. The Green's function can, however, also be expressed without making direct use of the band states, namely as

$$\mathbf{G}_{a,b}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu) \mathbf{S} - \mathbf{h} - \boldsymbol{\Sigma}(i\omega_\nu) \right]_{a,b}^{-1} \quad (\text{B.10})$$

where

$$S_{a,b}^{-1} = \sum_k \langle \pi_a | \psi_k \rangle f_k \langle \psi_k | \pi_b \rangle \quad (\text{B.11})$$

Proof: Here, I will show that the Green's function matrix

$$\mathbf{G}_{a,b}(i\omega_\nu) = \sum_n \langle \pi_a | \psi_n \rangle \left[(i\hbar\omega_\nu + \mu) \delta_{n,n'} - \sum_{c,d} \langle \psi_n | \pi_c \rangle (h_{c,d} + \Sigma_{c,d}(i\omega_\nu)) \langle \pi_d | \psi_{n'} \rangle \right]_{n,n'}^{-1} \langle \psi_{n'} | \pi_b \rangle \quad (\text{B.12})$$

can be expressed in the form of Eq. B.10.

In order to simplify the proof, I will not write out the self energy, nor the chemical potential. This is possible because the proof works for each Matsubara frequency independently, so that

both can be absorbed in the non-interacting Hamiltonian.

$$\begin{aligned}
G_{a,b}(i\omega_\nu) &= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left(i\hbar\omega_\nu \delta_{n,n'} - \langle \psi_n | \pi \rangle \mathbf{h} \langle \pi | \psi_{n'} \rangle \right)_{n,n'}^{-1} \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left[\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\langle \psi | \pi \rangle \mathbf{h} \langle \pi | \psi \rangle \right)_{n,n'}^j \right] \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left[\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\sum_{n_2, \dots, n_j} \langle \psi_n | \pi \rangle \mathbf{h} \langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle \dots \mathbf{h} \langle \pi | \psi_{n'} \rangle \right) \right] \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\sum_{n_1, \dots, n_{j+1}} \underbrace{\langle \pi | \psi_{n_1} \rangle \langle \psi_{n_1} | \pi \rangle}_{\mathbf{S}^{-1}} \mathbf{h} \underbrace{\langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle}_{\mathbf{S}^{-1}} \dots \mathbf{h} \underbrace{\langle \pi | \psi_{n_{j+1}} \rangle \langle \psi_{n_{j+1}} | \pi \rangle}_{\mathbf{S}^{-1}} \right)_{a,b} \\
&= \left(\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\mathbf{S}^{-1} \mathbf{h} \right)^j \mathbf{S}^{-1} \right)_{a,b} \\
&= \left[i\hbar\omega_\nu \mathbf{S} \left(\mathbf{1} - \frac{1}{i\hbar\omega_\nu} \mathbf{S}^{-1} \mathbf{h} \right) \right]_{a,b}^{-1} \\
&= \left[i\hbar\omega_\nu \mathbf{S} - \mathbf{h} \right]_{a,b}^{-1}
\end{aligned} \tag{B.13}$$

q.e.d.

B.2.1 Laurent expansion of the Green's function

Here, we extract the Laurent expansion of the Green's function in the form

$$\mathbf{G} = \left[(i\hbar\omega_\nu + \mu) \mathbf{S} - \mathbf{h} - \boldsymbol{\Sigma}(i\omega_\nu) \right]^{-1} \tag{B.14}$$

where the self energy has the Laurent expansion

$$\boldsymbol{\Sigma}(i\omega_\nu) = \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j} \mathcal{S}^{(j)} \tag{B.15}$$

$$\begin{aligned}
\mathbf{G} &= \left[(i\hbar\omega_\nu) \mathbf{S} \left(\mathbf{1} - \frac{1}{i\hbar\omega_\nu} \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \boldsymbol{\Sigma}(i\omega_\nu) \right) \right) \right]^{-1} \\
&= \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \boldsymbol{\Sigma}(i\omega_\nu) \right) \right)^j \mathbf{S}^{-1} \\
&= \frac{1}{(i\hbar\omega_\nu)} \mathbf{S}^{-1} + \frac{1}{(i\hbar\omega_\nu)^2} \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} + \frac{1}{i\hbar\omega_\nu} \mathcal{S}^{(1)} \right) \mathbf{S}^{-1} \\
&\quad + \frac{1}{(i\hbar\omega_\nu)^3} \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} + O(\omega_\nu^{-4}) \\
&= \frac{1}{(i\hbar\omega_\nu)} \mathbf{S}^{-1} + \frac{1}{(i\hbar\omega_\nu)^2} \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} \\
&\quad + \frac{1}{(i\hbar\omega_\nu)^3} \mathbf{S}^{-1} \left(\mathcal{S}^{(1)} + \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \right) \mathbf{S}^{-1} + O(\omega_\nu^{-4})
\end{aligned} \tag{B.16}$$

$$\mathbf{G} = \sum_{j=1}^{\infty} \frac{1}{(i\hbar\omega_{\nu})^j} \mathcal{G}^{(j)} \quad (\text{B.17})$$

with

$$\begin{aligned} \mathcal{G}^{(1)} &= \mathbf{S}^{-1} \\ \mathcal{G}^{(2)} &= \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} \\ \mathcal{G}^{(3)} &= \mathbf{S}^{-1} \left(\mathcal{S}^{(1)} + \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \mathbf{S}^{-1} \left(\mathbf{h} - \mu \mathbf{S} + \mathcal{S}^{(0)} \right) \right) \mathbf{S}^{-1} \end{aligned} \quad (\text{B.18})$$

The expansion coefficients for the Laurent expansion are hermitian. We exploit that $\mathbf{G}(-i\omega_{\nu}) = \mathbf{G}^{\dagger}(i\omega_{\nu})$

$$\begin{aligned} \sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_{\nu})^j} \mathcal{G}^{(j)} &= \mathbf{G}(-i\omega_{\nu}) = \mathbf{G}^{\dagger}(i\omega_{\nu}) = \sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_{\nu})^j} \left(\mathcal{G}^{(j)} \right)^{\dagger} \\ \Rightarrow \quad \mathcal{G}^{(j)} &= \left(\mathcal{G}^{(j)} \right)^{\dagger} \end{aligned} \quad (\text{B.19})$$

From this requirement we can derive that \mathbf{S} , \mathbf{h} , $\mathcal{S}^{(0)}$ and $\mathcal{S}^{(0)}$ are hermitian as well.

Laurent expansion for the density matrix

With this expression, we obtain the Matsubara sum required for the density matrix as

$$\begin{aligned} \frac{1}{\beta} \sum_{\nu} \mathbf{G} e^{i\omega_{\nu} 0^+} &= \sum_{j=1}^{\infty} \mathcal{G}^{(j)} \frac{1}{(j-1)!} \partial_{\epsilon}^{j-1} \Big|_{\epsilon=0} (1 + e^{\beta\epsilon})^{-1} \\ &= \frac{1}{2} \mathcal{G}^{(1)} - \frac{1}{4} \beta \mathcal{G}^{(2)} - 0 \cdot \beta^2 \mathcal{G}^{(3)} + \underbrace{\left(\frac{1}{3!} \frac{1}{8} \beta^3 \mathcal{G}^{(4)} - \frac{1}{5!} \frac{1}{4} \beta^5 \mathcal{G}^{(6)} + \frac{1}{7!} 1.0? \beta^7 \mathcal{G}^{(8)} \right)}_{\text{from grapher}} + O(\beta^9) \\ &= \frac{1}{2} \mathcal{G}^{(1)} - \frac{1}{4} \beta \mathcal{G}^{(2)} + \underbrace{\left(\frac{1}{48} \beta^3 \mathcal{G}^{(4)} - \frac{1}{480} \beta^5 \mathcal{G}^{(6)} + \frac{1.0?}{4320} \beta^7 \mathcal{G}^{(8)} \right)}_{\text{from grapher}} + O(\beta^9) \end{aligned}$$

B.3 Model Green's function

In order to analyze the equations it is often useful to have a model Green's function that has an analytical form but reflects the qualitative properties of the Green's function.

We start with the expression for a non-interacting Green's function in terms of Matsubara frequencies

$$G_{a,b}(i\omega_{\nu}) = \sum_n \frac{\langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle}{i\hbar\omega_{\nu} + \mu - \epsilon_n} = \int d\epsilon \frac{D_{a,b}(\epsilon)}{i\hbar\omega_{\nu} + \mu - \epsilon} \quad (\text{B.20})$$

Now we introduce a model density of states that is diagonal in the orbital index and which has a constant density of states of width W centered at the energy E

$$D_{a,b}(\epsilon) = \delta_{a,b} \frac{1}{W} \cdot \underbrace{\theta\left(\epsilon - \left(E - \frac{W}{2}\right)\right)}_{=1 \text{ above lower bound}} \cdot \underbrace{\theta\left(\left(E + \frac{W}{2}\right) - \epsilon\right)}_{=1 \text{ below upper bound}} \quad (\text{B.21})$$

The density of states is normalized so that the total weight of each index equals unity. The Heaviside function $\theta(x)$ is defined such that it vanishes for negative arguments and is equal to one for positive arguments.

We calculate the green's function for $\mu = 0$. The relative position of chemical potential and the band can be changed easily by adapting the position of the band, i.e. by changing E . Keeping E flexible rather than μ also expand the argument to multi-band situations. The resulting Green's function has the form

$$\begin{aligned} G_{a,b}(i\omega_\nu) &= \int_{E-W/2}^{E+W/2} d\epsilon \frac{\frac{1}{W}\delta_{a,b}}{i\hbar\omega_\nu - \epsilon} \\ &= \frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} d\epsilon \frac{-\epsilon - i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\ &= -\frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} d\epsilon \frac{\epsilon + i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\ &= -\frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} dx \frac{x + i\hbar\omega_\nu}{(\hbar\omega)^2 + x^2} \\ &= -\frac{\delta_{a,b}}{W} \left\{ \int_{E-W/2}^{E+W/2} dx \frac{x}{(\hbar\omega)^2 + x^2} + i\hbar\omega_\nu \int_{E-W/2}^{E+W/2} dx \frac{1}{(\hbar\omega)^2 + x^2} \right\} \\ &= -\frac{\delta_{a,b}}{W} \left\{ \frac{1}{2} \ln [(\hbar\omega)^2 + (E + W/2)^2] - \frac{1}{2} \ln [(\hbar\omega)^2 + (E - W/2)^2] \right. \\ &\quad \left. + i\hbar\omega_\nu \left[\frac{1}{\hbar\omega} \operatorname{atan} \left(\frac{E + W/2}{\hbar\omega} \right) - \frac{1}{\hbar\omega} \operatorname{atan} \left(\frac{E - W/2}{\hbar\omega} \right) \right] \right\} \\ &= \frac{\delta_{a,b}}{W} \left\{ +\frac{1}{2} \ln \left[\frac{(\hbar\omega)^2 + (E - W/2)^2}{(\hbar\omega)^2 + (E + W/2)^2} \right] - i \left[\operatorname{atan} \left(\frac{E + W/2}{\hbar\omega} \right) - \operatorname{atan} \left(\frac{E - W/2}{\hbar\omega} \right) \right] \right\} \end{aligned} \quad (\text{B.22})$$

We used the integral formula's from Bronstein¹

The result as function of ω is shown in Fig. B.1 The real part is symmetric about the origin, while the imaginary part is antisymmetric. This is consistent with $\mathbf{G}(-i\omega_\nu) = \mathbf{G}^\dagger(-i\omega_\nu)$.

The imaginary part exhibits a step at the origin from π/W to π/W for metallic systems, that is for a finite density of states at the origin. For an insulating system the imaginary part of the Green's function is smooth and has a finite slope at the origin.

¹Integral formula's:

$$\begin{aligned} \int dx \frac{1}{a^2 + x^2} &= \frac{1}{a} \operatorname{atan} \left(\frac{x}{a} \right) \\ \int dx \frac{x}{a^2 + x^2} &= \frac{1}{2} \ln (a^2 + x^2) \end{aligned} \quad (\text{B.23})$$

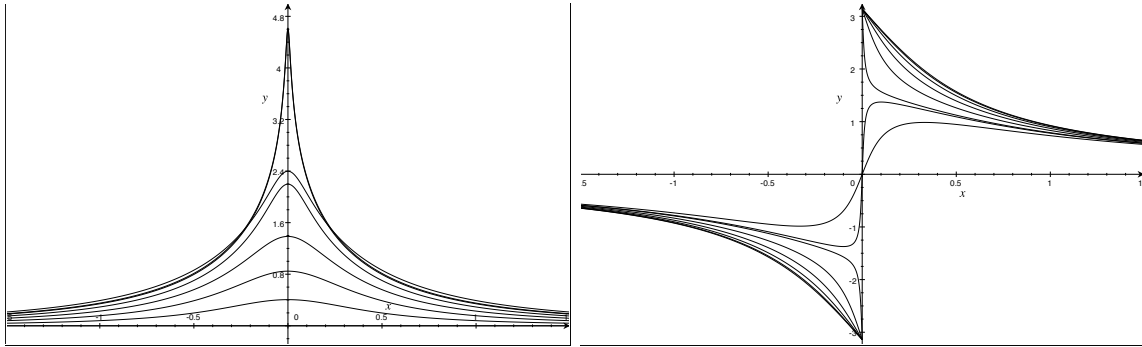


Fig. B.1: Negative real part (left) and negative imaginary part (right) of the Greens function with a constant density of states of width $W = 1$, centered at $E = \{0, 0.1, 0.2, 0.3, 0.49, 0.51, 0.6\}$.

The real part is positive if the band is centered above the chemical potential and it is negative if it is centered below. The real part vanishes if the density of states at the Fermi level. The real part becomes spiky exactly when the band touches the Fermi level. As the density of states shifts further to positive energies, the Greens function grows further in the tails, but it shrinks in the central region.

The following is not correct! Please check!

Limiting cases

- $\hbar\omega \rightarrow 0$:

We use the expansion of the arcus tangens ² and the expansion of the logarithm ³.

$$\begin{aligned}
G_{a,b}(i\omega_\nu) &= \frac{\delta_{a,b}}{W} \left\{ +\frac{1}{2} \ln \left[\frac{(\hbar\omega)^2 + (E - W/2)^2}{(\hbar\omega)^2 + (E + W/2)^2} \right] - i \left[\text{atan} \left(\frac{E + W/2}{\hbar\omega} \right) - \text{atan} \left(\frac{E - W/2}{\hbar\omega} \right) \right] \right\} \\
&\approx \frac{\delta_{a,b}}{W} \left\{ +\frac{1}{2} \ln \left[\left(\frac{E - W/2}{E + W/2} \right)^2 \right] + \frac{1}{2} \left[\frac{1}{(E - W/2)^2} - \frac{1}{(E + W/2)^2} \right] \hbar\omega^2 \right. \\
&\quad \left. - i \left[\frac{\pi}{2} - \frac{\hbar\omega}{E + W/2} - \underbrace{\frac{\pi}{2} + \frac{\hbar\omega}{E - W/2}}_{\text{times -1 for insulators}} \right] \right\} \\
&\approx \frac{\delta_{a,b}}{W} \left\{ +\ln \left[\frac{E - W/2}{E + W/2} \right] + \frac{EW}{\left(E^2 - \left(\frac{W}{2}\right)^2\right)^2} \hbar\omega^2 - i \left\{ \begin{array}{ll} \frac{W}{E^2 - \left(\frac{W}{2}\right)^2} \hbar\omega & \text{for insulators} \\ \pm\pi - \frac{4|E|}{E^2 - \left(\frac{W}{2}\right)^2} \hbar\omega & \text{for metals} \end{array} \right\} \right\}
\end{aligned} \tag{B.26}$$

• $\hbar\omega \rightarrow \infty$:

$$\begin{aligned}
G_{a,b}(i\omega_\nu) &= \frac{\delta_{a,b}}{W} \left\{ +\frac{1}{2} \ln \left[\frac{(\hbar\omega)^2 + (E - W/2)^2}{(\hbar\omega)^2 + (E + W/2)^2} \right] - i \left[\text{atan} \left(\frac{E + W/2}{\hbar\omega} \right) - \text{atan} \left(\frac{E - W/2}{\hbar\omega} \right) \right] \right\} \\
&\approx \frac{\delta_{a,b}}{W} \left\{ -EW(\hbar\omega)^{-2} - iW(\hbar\omega)^{-1} \right\} \\
&\approx \delta_{a,b} \left\{ -E(\hbar\omega)^{-2} - i(\hbar\omega)^{-1} \right\}
\end{aligned} \tag{B.27}$$

Remark: We can approximate

$$\frac{1}{G_{a,a}^2} \approx \left(\frac{1}{W} \ln \left[\frac{E - W/2}{E + W/2} \right] \right)^{-2} + (\hbar\omega)^2 \tag{B.28}$$

B.4 Bloch representation

2

$$\begin{aligned}
\text{atan}(x) = y &\Rightarrow y = \tan(x) = \frac{\sin(x)}{\cos(x)} \\
x = \frac{1}{\frac{\pi}{2} - y} + O(y - \frac{\pi}{2})^2 &\Rightarrow y \approx \frac{\pi}{2} - \frac{1}{x} \quad \text{for } x \rightarrow +\infty \\
x = \frac{1}{y + \frac{\pi}{2}} + O(y + \frac{\pi}{2})^2 &\Rightarrow y \approx -\frac{\pi}{2} + \frac{1}{x} \quad \text{for } x \rightarrow -\infty
\end{aligned} \tag{B.24}$$

3

$$\ln \left[\frac{a+x}{b+x} \right] \approx \ln \left[\frac{a}{b} \right] + \left(\frac{1}{a} - \frac{1}{b} \right) x + O(x^2) \tag{B.25}$$

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